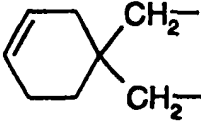
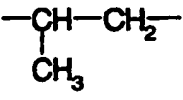




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(21) International Application Number: PCT/EP99/01820 (22) International Filing Date: 19 March 1999 (19.03.99) (30) Priority Data: 694/98 24 March 1998 (24.03.98) CH (71) Applicant (for all designated States except US): CIBA SPECIALTY CHEMICALS HOLDING INC. [CH/CH]; Klybeckstrasse 141, CH-4057 Basel (CH). (72) Inventors; and (75) Inventors/Applicants (for US only): KROEHNKE, Christoph [DE/DE]; Kleingasse 23, D-79206 Breisach (DE). DREWES, Rolf [DE/DE]; Im Mittelgrund 4, D-79415 Bad Bellingen (DE). (74) Common Representative: CIBA SPECIALTY CHEMICALS HOLDING INC.; Patentabteilung, Klybeckstrasse 141, CH-4057 Basel (CH).		(81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG). Published <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
(54) Title: STABILISATION OF ORGANIC MATERIALS (57) Abstract <p>Organic materials, especially polyolefins, having excellent stability against oxidative, thermal or light-induced degradation comprise, as stabilisers, (α) at least one compound of formula (A), wherein n' is a number from 1 to 6, R'_1, R'_2, R'_3 and R'_4 are each independently of the others C_4-C_{22}-alkyl, C_7-C_9phenylalkyl; or C_5-C_8cycloalkyl unsubstituted or substituted by C_1-C_4alkyl; or R'_1 and R'_2 together and/or R'_3 and R'_4 together are C_2-C_9alkylene or a group of formula (a), R'_5 and R'_6 are each independently of the other $-CH_2-CH_2-$ or formula (b), and (β) (i) at least one compound of the benzofuran-2-one type or (ii) α-tocopherol.</p> <div style="text-align: center;"> $R'_1-O-\left[\begin{array}{c} O-R'_2 \\ \\ P-O-R'_5-O-R'_6-O \\ \\ O-R'_3 \\ \\ O-R'_4 \end{array} \right]_{n'} \quad (A)$ </div> <div style="display: flex; justify-content: space-around; align-items: center;"> <div style="text-align: center;">  <p>(a)</p> </div> <div style="text-align: center;">  <p>(b)</p> </div> </div>		

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Stabilisation of organic materials

The present invention relates to compositions comprising an organic material subject to oxidative, thermal or light-induced degradation, especially polyolefin, and, as stabilisers, at least one monomeric or oligomeric organic bisphosphite and at least one compound of the benzofuran-2-one type or α -tocopherol (vitamin E), to the use of the same in stabilising organic materials, especially polyolefins, against oxidative, thermal or light-induced degradation, and to a method for stabilising those organic materials.

A large number of aliphatic bisphosphites derived from dihydric alcohols are known from the literature and are described, for example, in U.S. 3,342,767 or EP-A-0 635 511.

It is known, for example, from U.S. 5,516,920 that polypropylene can be protected against oxidative damage during processing by means of suitable stabiliser mixtures comprising certain aromatic phosphites or phosphonites and certain benzofuran-2-ones.

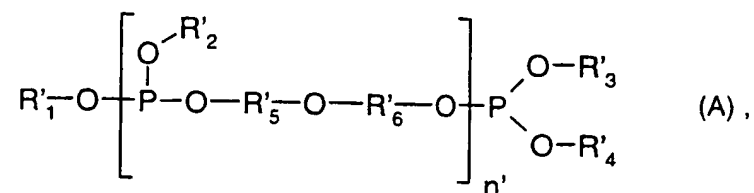
Those known stabiliser mixtures do not in every respect satisfy the high requirements to be met by a stabiliser mixture, especially as regards storage stability, water absorption, sensitivity to hydrolysis, stabilisation during processing, colour behaviour, volatility, migration behaviour, compatibility and enhanced light stabilisation. There is therefore still a need for effective stabilisers for organic materials, especially polyolefins, that are sensitive to oxidative, thermal or light-induced degradation.

It has now been found that a stabiliser mixture comprising at least one monomeric or oligomeric organic bisphosphite and at least one compound of the benzofuran-2-one type or α -tocopherol (vitamin E) is especially suitable as a stabiliser for organic materials, especially polyolefins, that are sensitive to oxidative, thermal or light-induced degradation.

The present invention accordingly relates to compositions comprising

- a) an organic material subject to oxidative, thermal or light-induced degradation,
- b) at least one compound of formula A

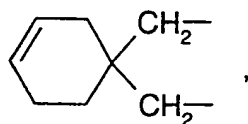
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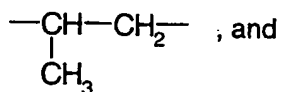
wherein

n' is a number from 1 to 6,

R'_1 , R'_2 , R'_3 and R'_4 are each independently of the others C_4 - C_{22} alkyl, C_7 - C_9 phenylalkyl; or C_5 - C_8 cycloalkyl unsubstituted or substituted by C_1 - C_4 alkyl; or R'_1 and R'_2 together and/or R'_3 and R'_4 together are C_2 - C_9 alkylene or a group of the formula



R'_5 and R'_6 are each independently of the other $-\text{CH}_2-\text{CH}_2-$ or



c) (i) at least one compound of the benzofuran-2-one type or (ii) α -tocopherol.

Alkyl having from 4 to 22 carbon atoms is a branched or unbranched radical, such as, for example, n-butyl, sec-butyl, isobutyl, tert-butyl, 2-ethylbutyl, n-pentyl, isopentyl, 1-methylpentyl, 1,3-dimethylbutyl, n-hexyl, 1-methylhexyl, n-heptyl, isoheptyl, 1,1,3,3-tetramethylbutyl, 1-methylheptyl, 3-methylheptyl, n-octyl, isooctyl, 2-ethylhexyl, 1,1,3-trimethylhexyl, 1,1,3,3-tetramethylpentyl, nonyl, isononyl, decyl, isodecyl, undecyl, isoundecyl, dodecyl, isododecyl, 1,1,3,3,5,5-hexamethylhexyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, icosyl or docosyl. One of the preferred definitions for R'_1 , R'_2 , R'_3 and R'_4 is, for example, C_6 - C_{22} alkyl, especially C_6 - C_{15} alkyl, e.g. C_8 - C_{12} alkyl.

C_7 - C_9 Phenylalkyl is, for example, benzyl, α -methylbenzyl, α,α -dimethylbenzyl or 2-phenylethyl. A preferred definition for R'_1 , R'_2 , R'_3 and R'_4 is, for example, benzyl.

C₅-C₈Cycloalkyl unsubstituted or substituted by C₁-C₄alkyl is, for example, cyclopentyl, methylcyclopentyl, dimethylcyclopentyl, cyclohexyl, methylcyclohexyl, dimethylcyclohexyl, trimethylcyclohexyl, tert-butylcyclohexyl, cycloheptyl or cyclooctyl. Preference is given to cyclohexyl and tert-butylcyclohexyl.

C₂-C₉Alkylene is a branched or unbranched radical, such as, for example, ethylene, propylene, trimethylene, tetramethylene, pentamethylene, hexamethylene, heptamethylene, octamethylene or nonamethylene. Preference is given to C₂-C₇alkylene, especially C₂-C₄alkylene, e.g. propylene.

Of interest are compositions comprising as component (b) a compound of formula A wherein n' is from 1 to 4.

Likewise of interest are compositions comprising as component (b) a compound of formula A wherein

R'₁, R'₂, R'₃ and R'₄ are each independently of the others C₆-C₂₂alkyl or C₇-C₉phenylalkyl; or R'₁ and R'₂ together and/or R'₃ and R'₄ together are C₂-C₇alkylene, and

R'₅ and R'₆ are each independently of the other $\text{—CH}_2\text{—CH}_2\text{—}$ or $\begin{array}{c} \text{—CH—CH}_2\text{—} \\ | \\ \text{CH}_3 \end{array}$.

Of special interest are compositions comprising as component (b) a compound of formula A wherein

R'₁, R'₂, R'₃ and R'₄ are each independently of the others C₆-C₁₅alkyl or C₇-C₉phenylalkyl; or R'₁ and R'₂ together and/or R'₃ and R'₄ together are C₂-C₄alkylene, and

R'₅ and R'₆ each is $\begin{array}{c} \text{—CH—CH}_2\text{—} \\ | \\ \text{CH}_3 \end{array}$.

Preference is given to compositions comprising as component (b) a compound of formula A wherein

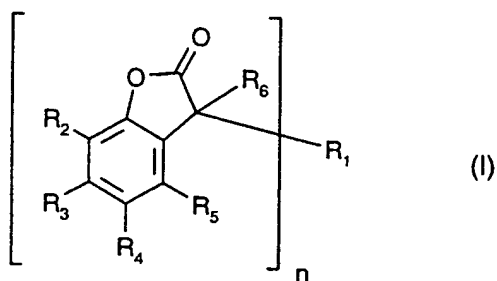
R'₁, R'₂, R'₃ and R'₄ are each independently of the others C₈-C₁₂alkyl, and

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R'₅ and R'₆ each is $\begin{array}{c} \text{---CH---CH}_2\text{---} \\ | \\ \text{CH}_3 \end{array}$

The compounds of formula A as component (b) in the composition according to the invention are known from the literature and the preparation thereof is described, for example, in U.S. 3,342,767 or EP-A-0 635 511.

Of interest are compositions comprising as component (c) (i) a compound of formula I

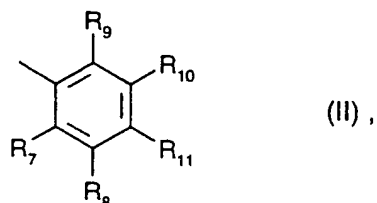


wherein,

when n is 1,

R₁ is naphthyl, phenanthryl, anthryl, 5,6,7,8-tetrahydro-2-naphthyl, 5,6,7,8-tetrahydro-1-naphthyl, thienyl, benzo[b]thienyl, naphtho[2,3-b]thienyl, thianthrenyl, dibenzofuryl, chromenyl, xanthenyl, phenoxathiinyl, pyrrolyl, imidazolyl, pyrazolyl, pyrazinyl, pyrimidinyl, pyridazinyl, indoliziny, isoindolyl, indolyl, indazolyl, purinyl, quinoliziny, isoquinolyl, quinolyl, phthalazinyl, naphthyridinyl, quinoxalinyl, quinazolinyl, cinnoliny, pteridinyl, carbazolyl, β-carboline, phenanthridinyl, acridinyl, perimidinyl, phenanthrolinyl, phenazinyl, isothiazolyl, phenothiazinyl, isoxazolyl, furazanyl, biphenyl, terphenyl, fluorenyl or phenoxazinyl each unsubstituted or substituted by C₁-C₄alkyl, C₁-C₄alkoxy, C₁-C₄alkylthio, hydroxy, halogen, amino, C₁-C₄alkylamino, phenylamino or by di(C₁-C₄alkyl)amino, or R₁ is a radical of formula II

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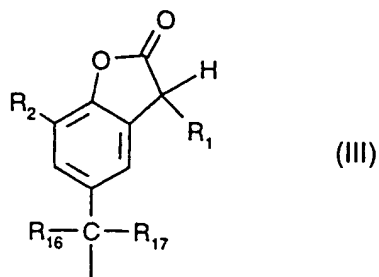
and,

when n is 2,

R_1 is phenylene or naphthylene each unsubstituted or substituted by C_1 - C_4 alkyl or by hydroxy; or $-R_{12}-X-R_{13}-$,

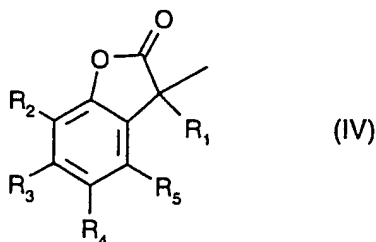
R_2 , R_3 , R_4 and R_5 are each independently of the others hydrogen, chlorine, hydroxy, C_1 - C_{25} -alkyl, C_7 - C_9 phenylalkyl; phenyl unsubstituted or substituted by C_1 - C_4 alkyl; C_5 - C_8 cycloalkyl unsubstituted or substituted by C_1 - C_4 alkyl; C_1 - C_{18} alkoxy, C_1 - C_{18} alkylthio, C_1 - C_4 alkylamino, di(C_1 - C_4 alkyl)amino, C_1 - C_{25} alkanoyloxy, C_1 - C_{25} alkanoylamino, C_3 - C_{25} -alkanoyloxy interrupted by oxygen, sulfur or by $\text{>N}-R_{14}$; C_6 - C_9 cycloalkylcarbonyloxy,

benzoyloxy, or benzoyloxy substituted by C_1 - C_{12} alkyl; or furthermore the radicals R_2 and R_3 or the radicals R_3 and R_4 or the radicals R_4 and R_5 , together with the carbon atoms to which they are bonded, form a benzo ring, in addition R_4 is $-(CH_2)_p-COR_{15}$ or $-(CH_2)_qOH$ or in addition, when R_3 , R_5 and R_6 are hydrogen, R_4 is a radical of formula III



wherein R_1 is defined as described above for when $n = 1$,

R_6 is hydrogen or a radical of formula IV



wherein R_4 is not a radical of formula III and R_1 is defined as described above for when $n = 1$,

R_7 , R_8 , R_9 , R_{10} and R_{11} are each independently of the others hydrogen, halogen, hydroxy,

C_1 - C_{25} alkyl; C_2 - C_{25} alkyl interrupted by oxygen, sulfur or by >N-R_{14} ; C_1 - C_{25} alkoxy;

C_2 - C_{25} alkoxy interrupted by oxygen, sulfur or by >N-R_{14} ; C_1 - C_{25} alkylthio, C_3 - C_{25} -

alkenyl, C_3 - C_{25} alkenyloxy, C_3 - C_{25} alkynyl, C_3 - C_{25} alkynyloxy, C_7 - C_9 phenylalkyl, C_7 - C_9 phenylalkoxy; phenyl unsubstituted or substituted by C_1 - C_4 alkyl; phenoxy unsubstituted or substituted by C_1 - C_4 alkyl; C_5 - C_8 cycloalkyl unsubstituted or substituted by C_1 - C_4 alkyl; C_5 - C_8 cycloalkoxy unsubstituted or substituted by C_1 - C_4 alkyl; C_1 - C_4 alkylamino, di(C_1 - C_4 alkyl)amino,

C_1 - C_{25} alkanoyl; C_3 - C_{25} alkanoyl interrupted by oxygen, sulfur or by >N-R_{14} ; C_1 - C_{25} -

alkanoyloxy; C_3 - C_{25} alkanoyloxy interrupted by oxygen, sulfur or by >N-R_{14} ; C_1 - C_{25} -

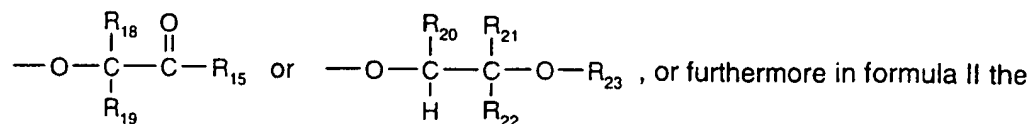
alkanoylamino, C_3 - C_{25} alkenoyl; C_3 - C_{25} alkenoyl interrupted by oxygen, sulfur or by

>N-R_{14} ; C_3 - C_{25} alkenoyloxy; C_3 - C_{25} alkenoyloxy interrupted by oxygen, sulfur or by

>N-R_{14} ; C_6 - C_9 cycloalkylcarbonyl, C_6 - C_9 cycloalkylcarbonyloxy, benzoyl, or benzoyl

substituted by C_1 - C_{12} alkyl; benzoyloxy, or benzoyloxy substituted by C_1 - C_{12} alkyl;

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radicals R_7 and R_8 or the radicals R_8 and R_{11} , together with the carbon atoms to which they are bonded, form a benzo ring,

R_{12} and R_{13} are each independently of the other phenylene or naphthylene each unsubstituted or substituted by C_1 - C_4 alkyl,

R_{14} is hydrogen or C_1 - C_8 alkyl,

R_{15} is hydroxy, $\left[-\text{O}^- \frac{1}{r} \text{M}^{r+} \right]$, C_1 - C_{18} alkoxy or $-\text{N} \begin{array}{l} \text{R}_{24} \\ \text{R}_{25} \end{array}$,

R_{16} and R_{17} are each independently of the other hydrogen, CF_3 , C_1 - C_{12} alkyl or phenyl, or

R_{16} and R_{17} , together with the carbon atom to which they are bonded, form a

C_5 - C_8 cycloalkylidene ring unsubstituted or substituted by from 1 to 3 C_1 - C_4 alkyl groups;

R_{18} and R_{19} are each independently of the other hydrogen, C_1 - C_4 alkyl or phenyl,

R_{20} is hydrogen or C_1 - C_4 alkyl,

R_{21} is hydrogen; phenyl unsubstituted or substituted by C_1 - C_4 alkyl; C_1 - C_{25} alkyl; C_2 - C_{25} alkyl

interrupted by oxygen, sulfur or by >N-R_{14} ; C_7 - C_9 phenylalkyl unsubstituted or substi-

tuted on the phenyl moiety by from 1 to 3 C_1 - C_4 alkyl groups; C_7 - C_{25} phenylalkyl interrupted

by oxygen, sulfur or by >N-R_{14} and unsubstituted or substituted on the phenyl moiety

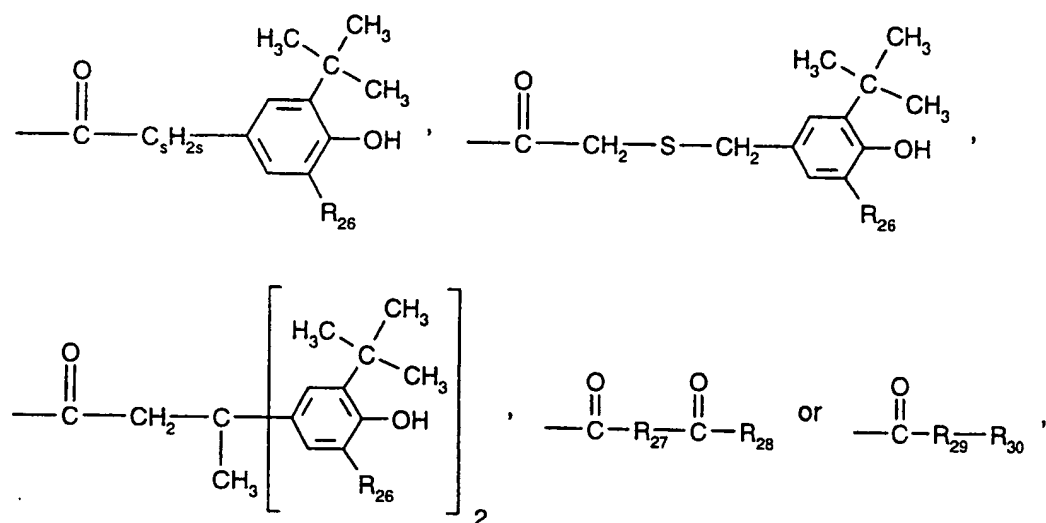
by from 1 to 3 C_1 - C_4 alkyl groups, or furthermore the radicals R_{20} and R_{21} , together with the carbon atoms to which they are bonded, form a C_5 - C_{12} cycloalkylene ring unsubstituted or substituted by from 1 to 3 C_1 - C_4 alkyl groups;

R_{22} is hydrogen or C_1 - C_4 alkyl,

R_{23} is hydrogen, C_1 - C_{25} alkanoyl, C_3 - C_{25} alkenoyl; C_3 - C_{25} alkanoyl interrupted by oxygen, sul-

fur or by >N-R_{14} ; C_2 - C_{25} alkanoyl substituted by a di(C_1 - C_6 alkyl) phosphonate group;

C_6 - C_9 cycloalkylcarbonyl, thenoyl, furoyl, benzoyl, or benzoyl substituted by C_1 - C_{12} alkyl;



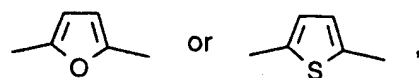
R₂₄ and R₂₅ are each independently of the other hydrogen or C₁-C₁₈alkyl,

R₂₆ is hydrogen or C₁-C₈alkyl,

R₂₇ is a direct bond, C₁-C₁₈alkylene; C₂-C₁₈alkylene interrupted by oxygen, sulfur or by



lene, C₇-C₈bicycloalkylene; phenylene unsubstituted or substituted by C₁-C₄alkyl;



R₂₈ is hydroxy, $\left[\text{---O}^- \frac{1}{r} \text{M}^{r+} \right]$, C₁-C₁₈alkoxy or $\text{---N(R}_{24}\text{)(R}_{25}\text{)---}$,

R₂₉ is oxygen, -NH- or $\text{---N(R}_{24}\text{)(R}_{25}\text{)---C(=O)---NH---R}_{30}$,

R₃₀ is C₁-C₁₈alkyl or phenyl,

R₃₁ is hydrogen or C₁-C₁₈alkyl,

M is an r-valent metal cation,

X is a direct bond, oxygen, sulfur or -NR₃₁- ,

n is 1 or 2,

p is 0, 1 or 2,

q is 1, 2, 3, 4, 5 or 6,

r is 1, 2 or 3, and

s is 0, 1 or 2.

Naphthyl, phenanthryl, anthryl, 5,6,7,8-tetrahydro-2-naphthyl, 5,6,7,8-tetrahydro-1-naphthyl, thienyl, benzo[b]thienyl, naphtho[2,3-b]thienyl, thianthrenyl, dibenzofuryl, chromenyl, xanthenyl, phenoxathiinyl, pyrrolyl, imidazolyl, pyrazolyl, pyrazinyl, pyrimidinyl, pyridazinyl, indoliziny, isoindolyl, indolyl, indazolyl, purinyl, quinoliziny, isoquinolyl, quinolyl, phthalazinyl, naphthyridinyl, quinoxaliny, quinazolinyl, cinnoliny, pteridinyl, carbazolyl, β -carboliny, phenanthridinyl, acridinyl, perimidinyl, phenanthrolinyl, phenazinyl, isothiazolyl, phenothiazinyl, isoxazolyl, furazanyl, biphenyl, terphenyl, fluorenyl or phenoxazinyl each unsubstituted or substituted by C₁-C₄alkyl, C₁-C₄alkoxy, C₁-C₄alkylthio, hydroxy, halogen, amino, C₁-C₄-alkylamino, phenylamino or by di(C₁-C₄alkyl)amino is, for example, 1-naphthyl, 2-naphthyl, 1-phenylamino-4-naphthyl, 1-methylnaphthyl, 2-methylnaphthyl, 1-methoxy-2-naphthyl, 2-methoxy-1-naphthyl, 1-dimethylamino-2-naphthyl, 1,2-dimethyl-4-naphthyl, 1,2-dimethyl-6-naphthyl, 1,2-dimethyl-7-naphthyl, 1,3-dimethyl-6-naphthyl, 1,4-dimethyl-6-naphthyl, 1,5-dimethyl-2-naphthyl, 1,6-dimethyl-2-naphthyl, 1-hydroxy-2-naphthyl, 2-hydroxy-1-naphthyl, 1,4-dihydroxy-2-naphthyl, 7-phenanthryl, 1-anthryl, 2-anthryl, 9-anthryl, 3-benzo[b]thienyl, 5-benzo[b]thienyl, 2-benzo[b]thienyl, 4-dibenzofuryl, 4,7-dibenzofuryl, 4-methyl-7-dibenzofuryl, 2-xanthenyl, 8-methyl-2-xanthenyl, 3-xanthenyl, 2-phenoxathiinyl, 2,7-phenoxathiinyl, 2-pyrrolyl, 3-pyrrolyl, 5-methyl-3-pyrrolyl, 2-imidazolyl, 4-imidazolyl, 5-imidazolyl, 2-methyl-4-imidazolyl, 2-ethyl-4-imidazolyl, 2-ethyl-5-imidazolyl, 3-pyrazolyl, 1-methyl-3-pyrazolyl, 1-propyl-4-pyrazolyl, 2-pyrazinyl, 5,6-dimethyl-2-pyrazinyl, 2-indoliziny, 2-methyl-3-isoindolyl, 2-methyl-1-isoindolyl, 1-methyl-2-indolyl, 1-methyl-3-indolyl, 1,5-dimethyl-2-indolyl, 1-methyl-3-indazolyl, 2,7-dimethyl-8-purinyl, 2-methoxy-7-methyl-8-purinyl, 2-quinoliziny, 3-isoquinolyl, 6-isoquinolyl, 7-isoquinolyl, isoquinolyl, 3-methoxy-6-isoquinolyl, 2-quinolyl, 6-quinolyl, 7-quinolyl, 2-methoxy-3-quinolyl, 2-methoxy-6-quinolyl, 6-phthalazinyl, 7-phthalazinyl, 1-methoxy-6-phthalazinyl, 1,4-dimethoxy-6-phthalazinyl, 1,8-naphthyridin-2-yl, 2-quinoxaliny, 6-quinoxaliny, 2,3-dimethyl-6-quinoxaliny, 2,3-dimethoxy-6-quinoxaliny, 2-quinazolinyl, 7-quinazolinyl, 2-dimethylamino-6-quinazolinyl, 3-cinnoliny, 6-cinnoliny, 7-cinnoliny, 3-methoxy-7-cinnoliny, 2-pteridinyl, 6-pteridinyl, 7-pteridinyl, 6,7-dimethoxy-2-pteridinyl, 2-carbazolyl, 3-carbazolyl, 9-methyl-2-carbazolyl, 9-methyl-3-carbazolyl, β -carbolin-3-yl, 1-methyl- β -carbolin-3-yl, 1-methyl- β -carbolin-6-yl, 3-phenanthridinyl, 2-acridinyl, 3-acridinyl, 2-perimidinyl, 1-methyl-5-perimidinyl, 5-phenanthrolinyl, 6-phenanthrolinyl, 1-phenazinyl, 2-phenazinyl, 3-isothiazolyl, 4-isothiazolyl, 5-isothiazolyl, 2-phenothiazinyl,

3-phenothiazinyl, 10-methyl-3-phenothiazinyl, 3-isoxazolyl, 4-isoxazolyl, 5-isoxazolyl, 4-methyl-3-furazanyl, 2-phenoxazinyl or 10-methyl-2-phenoxazinyl.

Special preference is given to naphthyl, phenanthryl, anthryl, 5,6,7,8-tetrahydro-2-naphthyl, 5,6,7,8-tetrahydro-1-naphthyl, thienyl, benzo[b]thienyl, naphtho[2,3-b]thienyl, thianthrenyl, dibenzofuryl, chromenyl, xanthenyl, phenoxathiinyl, pyrrolyl, isoindolyl, indolyl, phenothiazinyl, biphenyl, terphenyl, fluorenyl or phenoxazinyl each unsubstituted or substituted by C₁-C₄alkyl, C₁-C₄alkoxy, C₁-C₄alkylthio, hydroxy, phenylamino or by di(C₁-C₄alkyl)amino, such as, for example, 1-naphthyl, 2-naphthyl, 1-phenylamino-4-naphthyl, 1-methylnaphthyl, 2-methylnaphthyl, 1-methoxy-2-naphthyl, 2-methoxy-1-naphthyl, 1-dimethylamino-2-naphthyl, 1,2-dimethyl-4-naphthyl, 1,2-dimethyl-6-naphthyl, 1,2-dimethyl-7-naphthyl, 1,3-dimethyl-6-naphthyl, 1,4-dimethyl-6-naphthyl, 1,5-dimethyl-2-naphthyl, 1,6-dimethyl-2-naphthyl, 1-hydroxy-2-naphthyl, 2-hydroxy-1-naphthyl, 1,4-dihydroxy-2-naphthyl, 7-phenanthryl, 1-anthryl, 2-anthryl, 9-anthryl, 3-benzo[b]thienyl, 5-benzo[b]thienyl, 2-benzo[b]thienyl, 4-dibenzofuryl, 4,7-dibenzofuryl, 4-methyl-7-dibenzofuryl, 2-xanthenyl, 8-methyl-2-xanthenyl, 3-xanthenyl, 2-pyrrolyl, 3-pyrrolyl, 2-phenothiazinyl, 3-phenothiazinyl and 10-methyl-3-phenothiazinyl.

Halogen is, for example, chlorine, bromine or iodine. Preference is given to chlorine.

Alkanoyl having up to and including 25 carbon atoms is a branched or unbranched radical, such as, for example, formyl, acetyl, propionyl, butanoyl, pentanoyl, hexanoyl, heptanoyl, octanoyl, nonanoyl, decanoyl, undecanoyl, dodecanoyl, tridecanoyl, tetradecanoyl, pentadecanoyl, hexadecanoyl, heptadecanoyl, octadecanoyl, icosanoyl or docosanoyl. Preference is given to alkanoyl having from 2 to 18, especially from 2 to 12, e.g. from 2 to 6, carbon atoms. Special preference is given to acetyl.

C₂-C₂₅Alkanoyl substituted by a di(C₁-C₆alkyl) phosphonate group is, for example, (CH₃CH₂O)₂POCH₂CO-, (CH₃O)₂POCH₂CO-, (CH₃CH₂CH₂CH₂O)₂POCH₂CO-, (CH₃CH₂O)₂POCH₂CH₂CO-, (CH₃O)₂POCH₂CH₂CO-, (CH₃CH₂CH₂CH₂O)₂POCH₂CH₂CO-, (CH₃CH₂O)₂PO(CH₂)₄CO-, (CH₃CH₂O)₂PO(CH₂)₆CO- or (CH₃CH₂O)₂PO(CH₂)₁₇CO-.

Alkanoyloxy having up to and including 25 carbon atoms is a branched or unbranched radical, such as, for example, formyloxy, acetoxy, propionyloxy, butanoyloxy, pentanoyloxy, he-

xanoyloxy, heptanoyloxy, octanoyloxy, nonanoyloxy, decanoyloxy, undecanoyloxy, dodecanoyloxy, tridecanoyloxy, tetradecanoyloxy, pentadecanoyloxy, hexadecanoyloxy, heptadecanoyloxy, octadecanoyloxy, icosanoyloxy or docosanoyloxy. Preference is given to alkanoyloxy having from 2 to 18, especially from 2 to 12, e.g. from 2 to 6, carbon atoms. Special preference is given to acetoxy.

Alkenoyl having from 3 to 25 carbon atoms is a branched or unbranched radical, such as, for example, propenoyl, 2-butenoyl, 3-butenoyl, isobutenoyl, n-2,4-pentadienoyl, 3-methyl-2-butenoyl, n-2-octenoyl, n-2-dodecenoyl, isododecenoyl, oleoyl, n-2-octadecenoyl or n-4-octadecenoyl. Preference is given to alkenoyl having from 3 to 18, especially from 3 to 12, e.g. from 3 to 6, more especially 3 or 4, carbon atoms.

C₃-C₂₅Alkenoyl interrupted by oxygen, sulfur or by >N-R_{14} is, for example,

CH₃OCH₂CH₂CH=CHCO- or CH₃OCH₂CH₂OCH=CHCO-.

Alkenoyloxy having from 3 to 25 carbon atoms is a branched or unbranched radical, such as, for example, propenoyloxy, 2-butenoyloxy, 3-butenoyloxy, isobutenoyloxy, n-2,4-pentadienoyloxy, 3-methyl-2-butenoyloxy, n-2-octenoyloxy, n-2-dodecenoyloxy, isododecenoyloxy, oleoyloxy, n-2-octadecenoyloxy or n-4-octadecenoyloxy. Preference is given to alkenoyloxy having from 3 to 18, especially from 3 to 12, e.g. from 3 to 6, more especially 3 or 4, carbon atoms.

C₃-C₂₅Alkenoyloxy interrupted by oxygen, sulfur or by >N-R_{14} is, for example,

CH₃OCH₂CH₂CH=CHCOO- or CH₃OCH₂CH₂OCH=CHCOO-.

C₃-C₂₅Alkanoyl interrupted by oxygen, sulfur or by >N-R_{14} is, for example,

CH₃-O-CH₂CO-, CH₃-S-CH₂CO-, CH₃-NH-CH₂CO-, CH₃-N(CH₃)-CH₂CO-,
CH₃-O-CH₂CH₂-O-CH₂CO-, CH₃-(O-CH₂CH₂-)₂-O-CH₂CO-, CH₃-(O-CH₂CH₂-)₃-O-CH₂CO- or
CH₃-(O-CH₂CH₂-)₄-O-CH₂CO-.

C₃-C₂₅Alkanoyloxy interrupted by oxygen, sulfur or by >N-R_{14} is, for example,

CH₃-O-CH₂COO-, CH₃-S-CH₂COO-, CH₃-NH-CH₂COO-, CH₃-N(CH₃)-CH₂COO-,
 CH₃-O-CH₂CH₂-O-CH₂COO-, CH₃-(O-CH₂CH₂)₂O-CH₂COO-,
 CH₃-(O-CH₂CH₂)₃O-CH₂COO- or CH₃-(O-CH₂CH₂)₄O-CH₂COO-.

C₆-C₉Cycloalkylcarbonyl is, for example, cyclohexylcarbonyl, cycloheptylcarbonyl or cyclooctylcarbonyl. Preference is given to cyclohexylcarbonyl.

C₆-C₉Cycloalkylcarbonyloxy is, for example, cyclohexylcarbonyloxy, cycloheptylcarbonyloxy or cyclooctylcarbonyloxy. Preference is given to cyclohexylcarbonyloxy.

Benzoyl substituted by C₁-C₁₂alkyl, which benzoyl carries preferably from 1 to 3, especially 1 or 2, alkyl groups, is, for example, o-, m- or p-methylbenzoyl, 2,3-dimethylbenzoyl, 2,4-dimethylbenzoyl, 2,5-dimethylbenzoyl, 2,6-dimethylbenzoyl, 3,4-dimethylbenzoyl, 3,5-dimethylbenzoyl, 2-methyl-6-ethylbenzoyl, 4-tert-butylbenzoyl, 2-ethylbenzoyl, 2,4,6-trimethylbenzoyl, 2,6-dimethyl-4-tert-butylbenzoyl or 3,5-di-tert-butylbenzoyl. Preferred substituents are C₁-C₈alkyl, especially C₁-C₄alkyl.

Benzoyloxy substituted by C₁-C₁₂alkyl, which benzoyloxy carries preferably from 1 to 3, especially 1 or 2, alkyl groups, is, for example, o-, m- or p-methylbenzoyloxy, 2,3-dimethylbenzoyloxy, 2,4-dimethylbenzoyloxy, 2,5-dimethylbenzoyloxy, 2,6-dimethylbenzoyloxy, 3,4-dimethylbenzoyloxy, 3,5-dimethylbenzoyloxy, 2-methyl-6-ethylbenzoyloxy, 4-tert-butylbenzoyloxy, 2-ethylbenzoyloxy, 2,4,6-trimethylbenzoyloxy, 2,6-dimethyl-4-tert-butylbenzoyloxy or 3,5-di-tert-butylbenzoyloxy. Preferred substituents are C₁-C₈alkyl, especially C₁-C₄alkyl.

Alkyl having up to and including 25 carbon atoms is a branched or unbranched radical, such as, for example, methyl, ethyl, propyl, isopropyl, n-butyl, sec-butyl, isobutyl, tert-butyl, 2-ethylbutyl, n-pentyl, isopentyl, 1-methylpentyl, 1,3-dimethylbutyl, n-hexyl, 1-methylhexyl, n-heptyl, isoheptyl, 1,1,3,3-tetramethylbutyl, 1-methylheptyl, 3-methylheptyl, n-octyl, 2-ethylhexyl, 1,1,3-trimethylhexyl, 1,1,3,3-tetramethylpentyl, nonyl, decyl, undecyl, 1-methylundecyl, dodecyl, 1,1,3,3,5,5-hexamethylhexyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, icosyl or docosyl. One of the preferred definitions for R₂ and R₄ is, for example, C₁-C₁₈alkyl. An especially preferred definition for R₄ is C₁-C₄alkyl.

Alkenyl having from 3 to 25 carbon atoms is a branched or unbranched radical, such as, for example, propenyl, 2-butenyl, 3-butenyl, isobutenyl, n-2,4-pentadienyl, 3-methyl-2-butenyl, n-2-octenyl, n-2-dodecenyl, isododecenyl, oleyl, n-2-octadecenyl or n-4-octadecenyl. Preference is given to alkenyl having from 3 to 18, especially from 3 to 12, e.g. from 3 to 6, more especially 3 or 4, carbon atoms.

Alkenyloxy having from 3 to 25 carbon atoms is a branched or unbranched radical, such as, for example, propenyloxy, 2-butenyloxy, 3-butenyloxy, isobutenyloxy, n-2,4-pentadienyloxy, 3-methyl-2-butenyloxy, n-2-octenyloxy, n-2-dodecenyloxy, isododecenyloxy, oleyloxy, n-2-octadecenyloxy or n-4-octadecenyloxy. Preference is given to alkenyloxy having from 3 to 18, especially from 3 to 12, e.g. from 3 to 6, more especially 3 or 4, carbon atoms.

Alkynyl having from 3 to 25 carbon atoms is a branched or unbranched radical, such as, for example, propynyl ($\text{---CH}_2\text{---C}\equiv\text{CH}$), 2-butylnyl, 3-butylnyl, n-2-octynyl or n-2-dodecynyl. Preference is given to alkynyl having from 3 to 18, especially from 3 to 12, e.g. from 3 to 6, more especially 3 or 4, carbon atoms.

Alkynyloxy having from 3 to 25 carbon atoms is a branched or unbranched radical, such as, for example, propynyloxy ($\text{---OCH}_2\text{---C}\equiv\text{CH}$), 2-butyynyloxy, 3-butyynyloxy, n-2-octynyloxy or n-2-dodecynyloxy. Preference is given to alkynyloxy having from 3 to 18, especially from 3 to 12, e.g. from 3 to 6, more especially 3 or 4, carbon atoms.

$\text{C}_2\text{--C}_{25}$ Alkyl interrupted by oxygen, sulfur or by >N--R_{14} is, for example, $\text{CH}_3\text{--O--CH}_2\text{--}$,

$\text{CH}_3\text{--S--CH}_2\text{--}$, $\text{CH}_3\text{--NH--CH}_2\text{--}$, $\text{CH}_3\text{--N(CH}_3\text{)--CH}_2\text{--}$, $\text{CH}_3\text{--O--CH}_2\text{CH}_2\text{--O--CH}_2\text{--}$,
 $\text{CH}_3\text{--(O--CH}_2\text{CH}_2\text{--)}_2\text{O--CH}_2\text{--}$, $\text{CH}_3\text{--(O--CH}_2\text{CH}_2\text{--)}_3\text{O--CH}_2\text{--}$ or $\text{CH}_3\text{--(O--CH}_2\text{CH}_2\text{--)}_4\text{O--CH}_2\text{--}$.

$\text{C}_7\text{--C}_9$ Phenylalkyl is, for example, benzyl, α -methylbenzyl, α,α -dimethylbenzyl or 2-phenylethyl. Preference is given to benzyl and α,α -dimethylbenzyl.

C₇-C₉Phenylalkyl unsubstituted or substituted on the phenyl moiety by from 1 to 3 C₁-C₄alkyl groups is, for example, benzyl, α -methylbenzyl, α,α -dimethylbenzyl, 2-phenylethyl, 2-methylbenzyl, 3-methylbenzyl, 4-methylbenzyl, 2,4-dimethylbenzyl, 2,6-dimethylbenzyl or 4-tert-butylbenzyl. Preference is given to benzyl.

C₇-C₂₅Phenylalkyl interrupted by oxygen, sulfur or by >N-R_{14} and unsubstituted or substituted on the phenyl moiety by from 1 to 3 C₁-C₄alkyl groups is a branched or unbranched radical, such as, for example, phenoxymethyl, 2-methyl-phenoxymethyl, 3-methyl-phenoxymethyl, 4-methyl-phenoxymethyl, 2,4-dimethyl-phenoxymethyl, 2,3-dimethyl-phenoxymethyl, phenylthiomethyl, N-methyl-N-phenyl-aminomethyl, N-ethyl-N-phenyl-aminomethyl, 4-tert-butyl-phenoxymethyl, 4-tert-butyl-phenoxyethoxy-methyl, 2,4-di-tert-butyl-phenoxymethyl, 2,4-di-tert-butyl-phenoxyethoxymethyl, phenoxyethoxyethoxyethoxymethyl, benzyloxymethyl, benzyloxyethoxymethyl, N-benzyl-N-ethyl-aminomethyl or N-benzyl-N-isopropyl-aminomethyl.

C₇-C₉Phenylalkoxy is, for example, benzyloxy, α -methylbenzyloxy, α,α -dimethylbenzyloxy or 2-phenylethoxy. Preference is given to benzyloxy.

Phenyl substituted by C₁-C₄alkyl, which contains preferably from 1 to 3, especially 1 or 2, alkyl groups, is, for example, o-, m- or p-methylphenyl, 2,3-dimethylphenyl, 2,4-dimethylphenyl, 2,5-dimethylphenyl, 2,6-dimethylphenyl, 3,4-dimethylphenyl, 3,5-dimethylphenyl, 2-methyl-6-ethylphenyl, 4-tert-butylphenyl, 2-ethylphenyl or 2,6-diethylphenyl.

Phenoxy substituted by C₁-C₄alkyl, which contains preferably from 1 to 3, especially 1 or 2, alkyl groups, is, for example, o-, m- or p-methylphenoxy, 2,3-dimethylphenoxy, 2,4-dimethylphenoxy, 2,5-dimethylphenoxy, 2,6-dimethylphenoxy, 3,4-dimethylphenoxy, 3,5-dimethylphenoxy, 2-methyl-6-ethylphenoxy, 4-tert-butylphenoxy, 2-ethylphenoxy or 2,6-diethylphenoxy.

C₅-C₈Cycloalkyl unsubstituted or substituted by C₁-C₄alkyl is, for example, cyclopentyl, methylcyclopentyl, dimethylcyclopentyl, cyclohexyl, methylcyclohexyl, dimethylcyclohexyl, trimethylcyclohexyl, tert-butylcyclohexyl, cycloheptyl or cyclooctyl. Preference is given to cyclohexyl and tert-butylcyclohexyl.

C₅-C₈Cycloalkoxy unsubstituted or substituted by C₁-C₄alkyl is, for example, cyclopentyloxy, methylcyclopentyloxy, dimethylcyclopentyloxy, cyclohexyloxy, methylcyclohexyloxy, dimethylcyclohexyloxy, trimethylcyclohexyloxy, tert-butylcyclohexyloxy, cycloheptyloxy or cyclooctyloxy. Preference is given to cyclohexyloxy and tert-butylcyclohexyloxy.

Alkoxy having up to and including 25 carbon atoms is a branched or unbranched radical, such as, for example, methoxy, ethoxy, propoxy, isopropoxy, n-butoxy, isobutoxy, pentyloxy, isopentyloxy, hexyloxy, heptyloxy, octyloxy, decyloxy, tetradecyloxy, hexadecyloxy or octadecyloxy. Preference is given to alkoxy having from 1 to 12, especially from 1 to 8, e.g. from 1 to 6, carbon atoms.

C₂-C₂₅Alkoxy interrupted by oxygen, sulfur or by >N-R_{14} is, for example,

CH₃-O-CH₂CH₂O-, CH₃-S-CH₂CH₂O-, CH₃-NH-CH₂CH₂O-, CH₃-N(CH₃)-CH₂CH₂O-,
CH₃-O-CH₂CH₂-O-CH₂CH₂O-, CH₃-(O-CH₂CH₂)₂O-CH₂CH₂O-,
CH₃-(O-CH₂CH₂)₃O-CH₂CH₂O- or CH₃-(O-CH₂CH₂)₄O-CH₂CH₂O-.

Alkylthio having up to and including 25 carbon atoms is a branched or unbranched radical, such as, for example, methylthio, ethylthio, propylthio, isopropylthio, n-butylthio, isobutylthio, pentylthio, isopentylthio, hexylthio, heptylthio, octylthio, decylthio, tetradecylthio, hexadecylthio or octadecylthio. Preference is given to alkylthio having from 1 to 12, especially from 1 to 6, e.g. from 1 to 6, carbon atoms.

Alkylamino having up to and including 4 carbon atoms is a branched or unbranched radical, such as, for example, methylamino, ethylamino, propylamino, isopropylamino, n-butylamino, isobutylamino or tert-butylamino.

Di(C₁-C₄alkyl)amino means also that the two radicals are each independently of the other branched or unbranched, for example, dimethylamino, methylethylamino, diethylamino, methyl-n-propylamino, methylisopropylamino, methyl-n-butylamino, methylisobutylamino, ethylisopropylamino, ethyl-n-butylamino, ethylisobutylamino, ethyl-tert-butylamino, diethylamino, diisopropylamino, isopropyl-n-butylamino, isopropylisobutylamino, di-n-butylamino or di-isobutylamino.

Alkanoylamino having up to and including 25 carbon atoms is a branched or unbranched radical, such as, for example, formylamino, acetylamino, propionylamino, butanoylamino, pentanoylamino, hexanoylamino, heptanoylamino, octanoylamino, nonanoylamino, decanoylamino, undecanoylamino, dodecanoylamino, tridecanoylamino, tetradecanoylamino, pentadecanoylamino, hexadecanoylamino, heptadecanoylamino, octadecanoylamino, icosanoylamino or docosanoylamino. Preference is given to alkanoylamino having from 2 to 18, especially from 2 to 12, e.g. from 2 to 6, carbon atoms.

C₁-C₁₈Alkylene is a branched or unbranched radical, such as, for example, methylene, ethylene, propylene, trimethylene, tetramethylene, pentamethylene, hexamethylene, heptamethylene, octamethylene, decamethylene, dodecamethylene or octadecamethylene. Preference is given to C₁-C₁₂alkylene, especially C₁-C₈alkylene.

A C₅-C₁₂cycloalkylene ring unsubstituted or substituted by C₁-C₄alkyl, which contains preferably from 1 to 3, especially 1 or 2, branched or unbranched alkyl group radicals, is, for example, cyclopentylene, methylcyclopentylene, dimethylcyclopentylene, cyclohexylene, methylcyclohexylene, dimethylcyclohexylene, trimethylcyclohexylene, tert-butylcyclohexylene, cycloheptylene, cyclooctylene or cyclodecylene. Preference is given to cyclohexylene and tert-butylcyclohexylene.

C₂-C₁₈Alkylene interrupted by oxygen, sulfur or by >N-R_{14} is, for example,

-CH₂-O-CH₂-, -CH₂-S-CH₂-, -CH₂-NH-CH₂-, -CH₂-N(CH₃)-CH₂-, -CH₂-O-CH₂CH₂-O-CH₂-,
 -CH₂-(O-CH₂CH₂)₂O-CH₂-, -CH₂-(O-CH₂CH₂)₃O-CH₂-, -CH₂-(O-CH₂CH₂)₄O-CH₂- or
 -CH₂CH₂-S-CH₂CH₂-.

C₂-C₁₈Alkenylene is, for example, vinylene, methylvinylene, octenylethylene or dodecenylethylene. Preference is given to C₂-C₈alkenylene.

Alkylidene having from 2 to 20 carbon atoms is, for example, ethylidene, propylidene, butylidene, pentylidene, 4-methylpentylidene, heptylidene, nonylidene, tridecylidene, nonadecylidene, 1-methylethylidene, 1-ethylpropylidene or 1-ethylpentylidene. Preference is given to C₂-C₈alkylidene.

Phenylalkylidene having from 7 to 20 carbon atoms is, for example, benzylidene, 2-phenylethylidene or 1-phenyl-2-hexylidene. Preference is given to C₇-C₉phenylalkylidene.

C₅-C₈Cycloalkylene is a saturated hydrocarbon group having two free valences and at least one ring unit and is, for example, cyclopentylene, cyclohexylene, cycloheptylene or cyclooctylene. Preference is given to cyclohexylene.

C₇-C₈Bicycloalkylene is, for example, bicycloheptylene or bicyclooctylene.

Phenylene or naphthylene each unsubstituted or substituted by C₁-C₄alkyl is, for example, 1,2-, 1,3- or 1,4-phenylene or 1,2-, 1,3-, 1,4-, 1,6-, 1,7-, 2,6- or 2,7-naphthylene. Preference is given to 1,4-phenylene.

A C₅-C₈cycloalkylidene ring unsubstituted or substituted by C₁-C₄alkyl, which contains preferably from 1 to 3, especially 1 or 2, branched or unbranched alkyl group radicals, is, for example, cyclopentylidene, methylcyclopentylidene, dimethylcyclopentylidene, cyclohexylidene, methylcyclohexylidene, dimethylcyclohexylidene, trimethylcyclohexylidene, tert-butylcyclohexylidene, cycloheptylidene or cyclooctylidene. Preference is given to cyclohexylidene and tert-butylcyclohexylidene.

A mono-, di- or tri-valent metal cation is preferably an alkali metal cation, alkaline earth metal cation or aluminium cation, for example Na⁺, K⁺, Mg⁺⁺, Ca⁺⁺ or Al⁺⁺⁺.

Of interest are compositions comprising as component (c) (i) at least one compound of formula I wherein, when n is 1, R₁ is phenyl unsubstituted or substituted in the para-position by C₁-C₁₈alkylthio or by di(C₁-C₄alkyl)amino; mono- to penta-substituted alkylphenyl having an overall total of a maximum of 18 carbon atoms in the alkyl substituents, of which there are from 1 to 5; naphthyl, biphenyl, terphenyl, phenanthryl, anthryl, fluorenyl, carbazolyl, thienyl, pyrrolyl, phenothiazinyl or 5,6,7,8-tetrahydronaphthyl each unsubstituted or substituted by C₁-C₄alkyl, C₁-C₄alkoxy, C₁-C₄alkylthio, hydroxy or by amino.

Preference is given to compositions comprising as component (c) (i) at least one compound of formula I wherein, when n is 2,

R_1 is $-R_{12}-X-R_{13}-$,

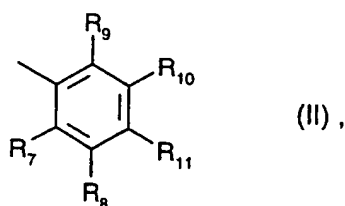
R_{12} and R_{13} each is phenylene,

X is oxygen or $-NR_{31}-$, and

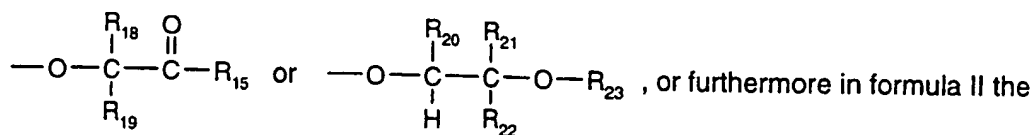
R_{31} is C_1-C_4 alkyl.

Preference is given also to compositions comprising as component (c) (i) at least one compound of formula I wherein, when n is 1,

R_1 is naphthyl, phenanthryl, thienyl, dibenzofuryl, carbazolyl or fluorenyl each unsubstituted or substituted by C_1-C_4 alkyl, C_1-C_4 alkoxy, C_1-C_4 alkylthio, hydroxy, halogen, amino, C_1-C_4 alkylamino or by $di(C_1-C_4$ alkyl)amino or is a radical of formula II



R_7 , R_8 , R_9 , R_{10} and R_{11} are each independently of the others hydrogen, chlorine, bromine, hydroxy, C_1-C_{18} alkyl; C_2-C_{18} alkyl interrupted by oxygen or by sulfur; C_1-C_{18} alkoxy; C_2-C_{18} alkoxy interrupted by oxygen or by sulfur; C_1-C_{18} alkylthio, C_3-C_{12} alkenyloxy, C_3-C_{12} alkynyloxy, C_7-C_9 phenylalkyl, C_7-C_9 phenylalkoxy; phenyl unsubstituted or substituted by C_1-C_4 alkyl; phenoxy, cyclohexyl, C_5-C_8 cycloalkoxy, C_1-C_4 alkylamino, $di(C_1-C_4$ alkyl)amino, C_1-C_{12} alkanoyl; C_3-C_{12} alkanoyl interrupted by oxygen or by sulfur; C_1-C_{12} alkanoyloxy; C_3-C_{12} alkanoyloxy interrupted by oxygen or by sulfur; C_1-C_{12} alkanoylamino, C_3-C_{12} alkenoyl, C_3-C_{12} alkenoyloxy, cyclohexylcarbonyl, cyclohexylcarbonyloxy, benzoyl, or benzoyl substituted by C_1-C_4 alkyl; benzoyloxy, or benzoyloxy substituted by C_1-C_4 alkyl;



radicals R_7 and R_8 or the radicals R_8 and R_{11} , together with the carbon atoms to which they are bonded, form a benzo ring,

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R_{15} is hydroxy, C_1 - C_{12} alkoxy or $\text{---N} \begin{matrix} R_{24} \\ R_{25} \end{matrix}$,

R_{18} and R_{19} are each independently of the other hydrogen or C_1 - C_4 alkyl,

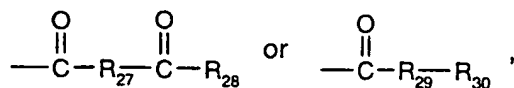
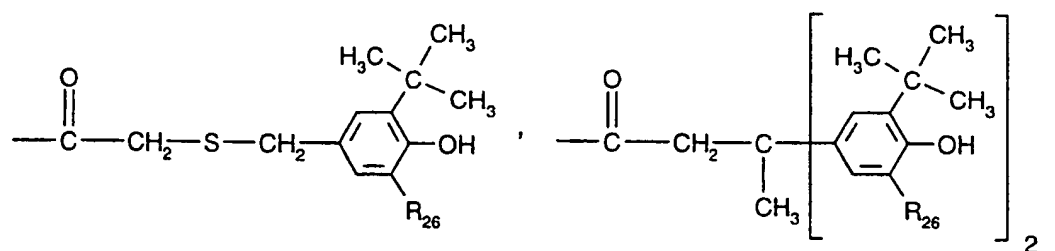
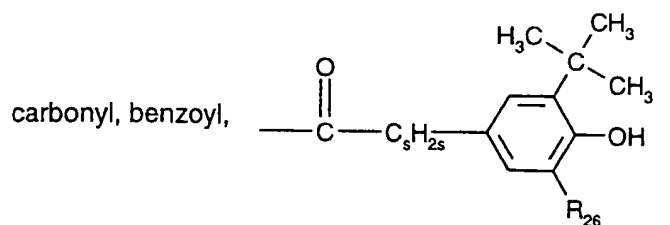
R_{20} is hydrogen,

R_{21} is hydrogen, phenyl, C_1 - C_{18} alkyl; C_2 - C_{18} alkyl interrupted by oxygen or by sulfur;

C_7 - C_9 phenylalkyl; C_7 - C_{18} phenylalkyl interrupted by oxygen or by sulfur and unsubstituted or substituted on the phenyl moiety by from 1 to 3 C_1 - C_4 alkyl groups, or furthermore the radicals R_{20} and R_{21} , together with the carbon atoms to which they are bonded, form a cyclohexylene ring unsubstituted or substituted by from 1 to 3 C_1 - C_4 alkyl groups,

R_{22} is hydrogen or C_1 - C_4 alkyl,

R_{23} is hydrogen, C_1 - C_{18} alkanoyl, C_3 - C_{18} alkenoyl; C_3 - C_{12} alkanoyl interrupted by oxygen or by sulfur; C_2 - C_{12} alkanoyl substituted by a di(C_1 - C_6 alkyl) phosphonate group; C_6 - C_9 cycloalkyl-



R_{24} and R_{25} are each independently of the other hydrogen or C_1 - C_{12} alkyl,

R_{26} is hydrogen or C_1 - C_4 alkyl,

R_{27} is C_1 - C_{12} alkylene, C_2 - C_8 alkenylene, C_2 - C_8 alkylidene, C_7 - C_{12} phenylalkylidene,

C_5 - C_8 cycloalkylene or phenylene,

R_{28} is hydroxy, C_1 - C_{12} alkoxy or $\text{---N} \begin{matrix} \nearrow R_{24} \\ \searrow R_{25} \end{matrix}$,

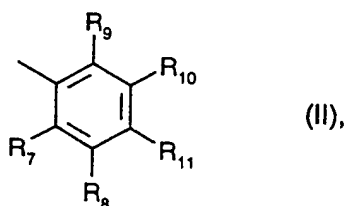
R_{29} is oxygen or ---NH--- ,

R_{30} is C_1 - C_{18} alkyl or phenyl, and

s is 1 or 2.

Preference is given also to compositions comprising as component (c) (i) at least one compound of formula I wherein, when n is 1,

R_1 is phenanthryl, thienyl, dibenzofuryl; carbazolyl unsubstituted or substituted by C_1 - C_4 alkyl; or fluorenyl; or R_1 is a radical of formula II



R_7 , R_8 , R_9 , R_{10} and R_{11} are each independently of the others hydrogen, chlorine, hydroxy, C_1 - C_{18} alkyl, C_1 - C_{18} alkoxy, C_1 - C_{18} alkylthio, C_3 - C_4 alkenyloxy, C_3 - C_4 alkynyloxy, phenyl,

benzoyl, benzoyloxy or $\text{---O---} \begin{matrix} R_{20} & R_{21} \\ | & | \\ \text{---C---} & \text{---C---} \\ | & | \\ H & R_{22} \end{matrix} \text{---O---} R_{23}$,

R_{20} is hydrogen,

R_{21} is hydrogen, phenyl or C_1 - C_{18} alkyl, or furthermore the radicals R_{20} and R_{21} , together with the carbon atoms to which they are bonded, form a cyclohexylene ring unsubstituted or substituted by from 1 to 3 C_1 - C_4 alkyl groups,

R_{22} is hydrogen or C_1 - C_4 alkyl, and

R_{23} is hydrogen, C_1 - C_{12} alkanoyl or benzoyl.

Special preference is given to compositions comprising as component (c) (i) at least one compound of formula I wherein, when n is 1,

R_7 , R_8 , R_9 , R_{10} and R_{11} are each independently of the others hydrogen, C_1 - C_{12} alkyl, C_1 - C_4 alkylthio or phenyl.

Of special interest are compositions comprising as component (c) (i) at least one compound of formula I wherein

R_2 , R_3 , R_4 and R_5 are each independently of the others hydrogen, chlorine, C_1 - C_{18} alkyl, benzyl, phenyl, C_5 - C_8 cycloalkyl, C_1 - C_{18} alkoxy, C_1 - C_{18} alkylthio, C_1 - C_{18} alkanoyloxy, C_1 - C_{18} -alkanoylamino, C_3 - C_{18} alkenoyloxy or benzoyloxy; or furthermore the radicals R_2 and R_3 or the radicals R_3 and R_4 or the radicals R_4 and R_5 , together with the carbon atoms to which they are bonded, form a benzo ring, in addition R_4 is $-(CH_2)_p-COR_{15}$ or $-(CH_2)_qOH$ or in addition, when R_3 , R_5 and R_6 are hydrogen, R_4 is a radical of formula III,

R_{15} is hydroxy, C_1 - C_{12} alkoxy or $\text{---N} \begin{matrix} \nearrow R_{24} \\ \searrow R_{25} \end{matrix}$,

R_{16} and R_{17} are methyl groups or, together with the carbon atom to which they are bonded, form a C_5 - C_8 cycloalkylidene ring unsubstituted or substituted by from 1 to 3 C_1 - C_4 alkyl groups,

R_{24} and R_{25} are each independently of the other hydrogen or C_1 - C_{12} alkyl,

p is 1 or 2, and

q is 2, 3, 4, 5 or 6.

Also of special interest are compositions comprising as component (c) (i) at least one compound of formula I wherein at least two of the radicals R_2 , R_3 , R_4 and R_5 are hydrogen.

Of special interest are, especially, compositions comprising as component (c) (i) at least one compound of formula I wherein R_3 and R_5 are hydrogen.

Of special interest are, more especially, compositions comprising as component (c) (i) at least one compound of formula I wherein

R_2 is C_1 - C_4 alkyl,

R_3 is hydrogen,

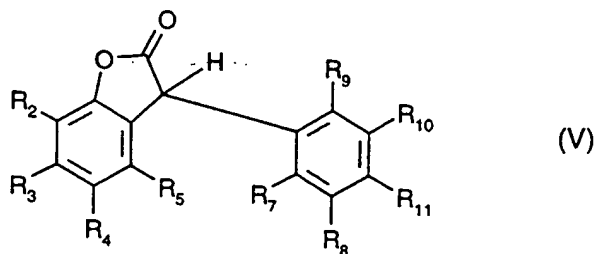
R_4 is C_1 - C_4 alkyl or in addition, when R_6 is hydrogen, R_4 is a radical of formula III,

R_5 is hydrogen, and

R₁₆ and R₁₇, together with the carbon atom to which they are bonded, form a cyclohexylidene ring.

The following compounds are examples of the benzofuran-2-one type that are especially suitable as component (c) (i) in the composition according to the invention: 3-[4-(2-acetoxyethoxy)phenyl]-5,7-di-tert-butyl-benzofuran-2-one; 5,7-di-tert-butyl-3-[4-(2-stearoyloxyethoxy)phenyl]-benzofuran-2-one; 3,3'-bis[5,7-di-tert-butyl-3-(4-[2-hydroxyethoxy]phenyl)-benzofuran-2-one]; 5,7-di-tert-butyl-3-(4-ethoxyphenyl)benzofuran-2-one; 3-(4-acetoxy-3,5-dimethylphenyl)-5,7-di-tert-butyl-benzofuran-2-one; 3-(3,5-dimethyl-4-pivaloyloxyphenyl)-5,7-di-tert-butyl-benzofuran-2-one; 5,7-di-tert-butyl-3-phenyl-benzofuran-2-one; 5,7-di-tert-butyl-3-(3,4-dimethylphenyl)-benzofuran-2-one; 5,7-di-tert-butyl-3-(2,3-dimethylphenyl)-benzofuran-2-one.

Also of special interest are, especially, compositions comprising as component (c) (i) at least one compound of formula V



wherein

R₂ is hydrogen or C₁-C₆alkyl,

R₃ is hydrogen,

R₄ is hydrogen or C₁-C₆alkyl,

R₅ is hydrogen,

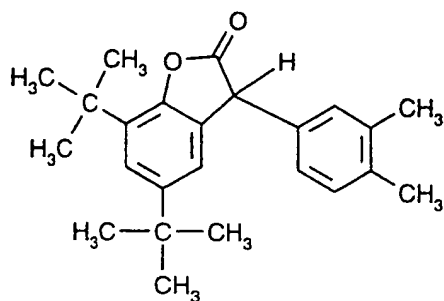
R₇, R₈, R₉, R₁₀ and R₁₁ are each independently of the others hydrogen, C₁-C₄alkyl, C₁-C₄-

alkoxy or $\text{—O—}\overset{\overset{\text{R}_{20}}{\text{|}}}{\underset{\underset{\text{H}}{\text{|}}}{\text{C}}}\text{—}\overset{\overset{\text{R}_{21}}{\text{|}}}{\underset{\underset{\text{R}_{22}}{\text{|}}}{\text{C}}}\text{—O—R}_{23}$, with the proviso that at least two of the radicals R₇,

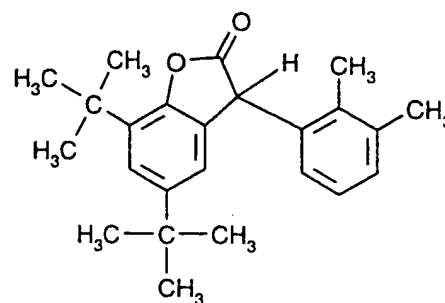
R₈, R₉, R₁₀ and R₁₁ are hydrogen,

R_{20} , R_{21} and R_{22} are hydrogen, and
 R_{23} is C_2 - C_4 alkanoyl.

Very special preference is given to compositions comprising as component (c) (i) at least one compound of formula Va or Vb



(Va)



(Vb)

or a mixture of both compounds of formulae Va and Vb.

The compounds of the benzofuran-2-one type as component (c) (i) in the composition according to the invention are known from the literature and the preparation thereof is described, for example, in U.S. 5,516,920.

The mixture of components (b) and (c) is suitable for stabilising organic materials against oxidative, thermal or light-induced degradation. Special attention is drawn to their excellent action as antioxidants in the stabilisation of organic materials.

1. Polymers of monoolefins and diolefins, for example polypropylene, polyisobutylene, polybut-1-ene, poly-4-methylpent-1-ene, polyisoprene or polybutadiene, as well as polymers of cycloolefins, for instance of cyclopentene or norbornene, polyethylene (which optionally can be crosslinked), for example high density polyethylene (HDPE), high density and high molecular weight polyethylene (HDPE-HMW), high density and ultrahigh molecular weight polyethylene (HDPE-UHMW), medium density polyethylene (MDPE), low density polyethylene (LDPE), linear low density polyethylene (LLDPE), (VLDPE) and (ULDPE).

Polyolefins, i.e. the polymers of monoolefins exemplified in the preceding paragraph, preferably polyethylene and polypropylene, can be prepared by different, and especially by the following, methods:

- a) radical polymerisation (normally under high pressure and at elevated temperature).
 - b) catalytic polymerisation using a catalyst that normally contains one or more than one metal of groups IVb, Vb, VIb or VIII of the Periodic Table. These metals usually have one or more than one ligand, typically oxides, halides, alcoholates, esters, ethers, amines, alkyls, alkenyls and/or aryls that may be either π - or σ -coordinated. These metal complexes may be in the free form or fixed on substrates, typically on activated magnesium chloride, titanium(III) chloride, alumina or silicon oxide. These catalysts may be soluble or insoluble in the polymerisation medium. The catalysts can be used by themselves in the polymerisation or further activators may be used, typically metal alkyls, metal hydrides, metal alkyl halides, metal alkyl oxides or metal alkyloxanes, said metals being elements of groups Ia, IIa and/or IIIa of the Periodic Table. The activators may be modified conveniently with further ester, ether, amine or silyl ether groups. These catalyst systems are usually termed Phillips, Standard Oil Indiana, Ziegler (-Natta), TNZ (DuPont), metallocene or single site catalysts (SSC).
2. Mixtures of the polymers mentioned under 1), for example mixtures of polypropylene with polyisobutylene, polypropylene with polyethylene (for example PP/HDPE, PP/LDPE) and mixtures of different types of polyethylene (for example LDPE/HDPE).
 3. Copolymers of monoolefins and diolefins with each other or with other vinyl monomers, for example ethylene/propylene copolymers, linear low density polyethylene (LLDPE) and mixtures thereof with low density polyethylene (LDPE), propylene/but-1-ene copolymers, propylene/isobutylene copolymers, ethylene/but-1-ene copolymers, ethylene/hexene copolymers, ethylene/methylpentene copolymers, ethylene/heptene copolymers, ethylene/octene copolymers, propylene/butadiene copolymers, isobutylene/isoprene copolymers, ethylene/alkyl acrylate copolymers, ethylene/alkyl methacrylate copolymers, ethylene/vinyl acetate copolymers and their copolymers with carbon monoxide or ethylene/acrylic acid copolymers and their salts (ionomers) as well as terpolymers of ethylene with propylene and a

diene such as hexadiene, dicyclopentadiene or ethylidene-norbornene; and mixtures of such copolymers with one another and with polymers mentioned in 1) above, for example polypropylene/ethylene-propylene copolymers, LDPE/ethylene-vinyl acetate copolymers (EVA), LDPE/ethylene-acrylic acid copolymers (EAA), LLDPE/EVA, LLDPE/EAA and alternating or random polyalkylene/carbon monoxide copolymers and mixtures thereof with other polymers, for example polyamides.

4. Hydrocarbon resins (for example C_5 - C_9) including hydrogenated modifications thereof (e.g. tackifiers) and mixtures of polyalkylenes and starch.

5. Polystyrene, poly(p-methylstyrene), poly(α -methylstyrene).

6. Copolymers of styrene or α -methylstyrene with dienes or acrylic derivatives, for example styrene/butadiene, styrene/acrylonitrile, styrene/alkyl methacrylate, styrene/butadiene/alkyl acrylate, styrene/butadiene/alkyl methacrylate, styrene/maleic anhydride, styrene/acrylonitrile/methyl acrylate; mixtures of high impact strength of styrene copolymers and another polymer, for example a polyacrylate, a diene polymer or an ethylene/propylene/diene terpolymer; and block copolymers of styrene such as styrene/butadiene/styrene, styrene/isoprene/styrene, styrene/ethylene/butylene/styrene or styrene/ethylene/propylene/styrene.

7. Graft copolymers of styrene or α -methylstyrene, for example styrene on polybutadiene, styrene on polybutadiene-styrene or polybutadiene-acrylonitrile copolymers; styrene and acrylonitrile (or methacrylonitrile) on polybutadiene; styrene, acrylonitrile and methyl methacrylate on polybutadiene; styrene and maleic anhydride on polybutadiene; styrene, acrylonitrile and maleic anhydride or maleimide on polybutadiene; styrene and maleimide on polybutadiene; styrene and alkyl acrylates or methacrylates on polybutadiene; styrene and acrylonitrile on ethylene/propylene/diene terpolymers; styrene and acrylonitrile on polyalkyl acrylates or polyalkyl methacrylates, styrene and acrylonitrile on acrylate/butadiene copolymers, as well as mixtures thereof with the copolymers listed under 6), for example the copolymer mixtures known as ABS, MBS, ASA or AES polymers.

8. Halogen-containing polymers such as polychloroprene, chlorinated rubbers, chlorinated and brominated copolymer of isobutylene-isoprene (halobutyl rubber), chlorinated or sulfochlorinated polyethylene, copolymers of ethylene and chlorinated ethylene, epichlorohydrin

homo- and copolymers, especially polymers of halogen-containing vinyl compounds, for example polyvinyl chloride, polyvinylidene chloride, polyvinyl fluoride, polyvinylidene fluoride, as well as copolymers thereof such as vinyl chloride/vinylidene chloride, vinyl chloride/vinyl acetate or vinylidene chloride/vinyl acetate copolymers.

9. Polymers derived from α,β -unsaturated acids and derivatives thereof such as polyacrylates and polymethacrylates; polymethyl methacrylates, polyacrylamides and polyacrylonitriles, impact-modified with butyl acrylate.
10. Copolymers of the monomers mentioned under 9) with each other or with other unsaturated monomers, for example acrylonitrile/ butadiene copolymers, acrylonitrile/alkyl acrylate copolymers, acrylonitrile/alkoxyalkyl acrylate or acrylonitrile/vinyl halide copolymers or acrylonitrile/ alkyl methacrylate/butadiene terpolymers.
11. Polymers derived from unsaturated alcohols and amines or the acyl derivatives or acetals thereof, for example polyvinyl alcohol, polyvinyl acetate, polyvinyl stearate, polyvinyl benzoate, polyvinyl maleate, polyvinyl butyral, polyallyl phthalate or polyallyl melamine; as well as their copolymers with olefins mentioned in 1) above.
12. Homopolymers and copolymers of cyclic ethers such as polyalkylene glycols, polyethylene oxide, polypropylene oxide or copolymers thereof with bisglycidyl ethers.
13. Polyacetals such as polyoxymethylene and those polyoxymethylenes which contain ethylene oxide as a comonomer; polyacetals modified with thermoplastic polyurethanes, acrylates or MBS.
14. Polyphenylene oxides and sulfides, and mixtures of polyphenylene oxides with styrene polymers or polyamides.
15. Polyurethanes derived from hydroxyl-terminated polyethers, polyesters or polybutadienes on the one hand and aliphatic or aromatic polyisocyanates on the other, as well as precursors thereof.

16. Polyamides and copolyamides derived from diamines and dicarboxylic acids and/or from aminocarboxylic acids or the corresponding lactams, for example polyamide 4, polyamide 6, polyamide 6/6, 6/10, 6/9, 6/12, 4/6, 12/12, polyamide 11, polyamide 12, aromatic polyamides starting from m-xylene diamine and adipic acid; polyamides prepared from hexamethylenediamine and isophthalic or/and terephthalic acid and with or without an elastomer as modifier, for example poly-2,4,4-trimethylhexamethylene terephthalamide or poly-m-phenylene isophthalamide; and also block copolymers of the aforementioned polyamides with polyolefins, olefin copolymers, ionomers or chemically bonded or grafted elastomers; or with polyethers, e.g. with polyethylene glycol, polypropylene glycol or polytetramethylene glycol; as well as polyamides or copolyamides modified with EPDM or ABS; and polyamides condensed during processing (RIM polyamide systems).
17. Polyureas, polyimides, polyamide-imides, polyetherimids, polyesterimids, polyhydantoins and polybenzimidazoles.
18. Polyesters derived from dicarboxylic acids and diols and/or from hydroxycarboxylic acids or the corresponding lactones, for example polyethylene terephthalate, polybutylene terephthalate, poly-1,4-dimethylolcyclohexane terephthalate and polyhydroxybenzoates, as well as block copolyether esters derived from hydroxyl-terminated polyethers; and also polyesters modified with polycarbonates or MBS.
19. Polycarbonates and polyester carbonates.
20. Polysulfones, polyether sulfones and polyether ketones.
21. Crosslinked polymers derived from aldehydes on the one hand and phenols, ureas and melamines on the other hand, such as phenol/formaldehyde resins, urea/formaldehyde resins and melamine/formaldehyde resins.
22. Drying and non-drying alkyd resins.
23. Unsaturated polyester resins derived from copolyesters of saturated and unsaturated dicarboxylic acids with polyhydric alcohols and vinyl compounds as crosslinking agents, and also halogen-containing modifications thereof of low flammability.

24. Crosslinkable acrylic resins derived from substituted acrylates, for example epoxy acrylates, urethane acrylates or polyester acrylates.
25. Alkyd resins, polyester resins and acrylate resins crosslinked with melamine resins, urea resins, isocyanates, isocyanurates, polyisocyanates or epoxy resins.
26. Crosslinked epoxy resins derived from aliphatic, cycloaliphatic, heterocyclic or aromatic glycidyl compounds, e.g. products of diglycidyl ethers of bisphenol A and bisphenol F, which are crosslinked with customary hardeners such as anhydrides or amines, with or without accelerators.
27. Natural polymers such as cellulose, rubber, gelatin and chemically modified homologous derivatives thereof, for example cellulose acetates, cellulose propionates and cellulose butyrates, or the cellulose ethers such as methyl cellulose; as well as rosins and their derivatives.
28. Blends of the aforementioned polymers (polyblends), for example PP/EPDM, Polyamide/EPDM or ABS, PVC/EVA, PVC/ABS, PVC/MBS, PC/ABS, PBTP/ABS, PC/ASA, PC/PBT, PVC/CPE, PVC/acrylates, POM/thermoplastic PUR, PC/thermoplastic PUR, POM/acrylate, POM/MBS, PPO/HIPS, PPO/PA 6.6 and copolymers, PA/HDPE, PA/PP, PA/PPO, PBT/PC/ABS or PBT/PET/PC.
29. Naturally occurring and synthetic organic materials which are pure monomeric compounds or mixtures of such compounds, for example mineral oils, animal and vegetable fats, oil and waxes, or oils, fats and waxes based on synthetic esters (e.g. phthalates, adipates, phosphates or trimellitates) and also mixtures of synthetic esters with mineral oils in any weight ratios, typically those used as spinning compositions, as well as aqueous emulsions of such materials.
30. Aqueous emulsions of natural or synthetic rubber, e.g. natural latex or latices of carboxylated styrene/butadiene copolymers.

Preferred organic materials are polymers, for example synthetic polymers, especially thermoplastic polymers. Especially preferred organic materials are polyolefins, polyurethanes and styrene copolymers, for example those mentioned above under points 1 to 3 and under points 6, 7 and 15, especially polyethylene and polypropylene and also ABS and styrene-butadiene copolymers.

Preferred organic materials are polyolefins, especially polyethylene or polypropylene and copolymers thereof with mono- and diolefins.

Special emphasis should be given to the action of the mixture of components (b) and (c) against oxidative and thermal degradation of polyolefins such as occurs in the processing of thermoplastics. Components (b) and (c) according to the invention are therefore excellent for use as processing stabilisers.

Preferably, component (b) is added to the organic material to be stabilised in an amount of from 0.01 to 10 %, for example from 0.01 to 5 %, preferably from 0.025 to 3 %, especially from 0.025 to 1 %, based on the weight of the organic material to be stabilised.

Advantageously, component (c) (i) is added to the organic material to be stabilised in an amount of from 0.0005 to 5 %, especially from 0.001 to 2 %, for example from 0.01 to 2 %, based on the weight of the organic material to be stabilised.

Advantageously, component (c) (ii) is added to the organic material to be stabilised in an amount of from 0.005 to 2 %, especially from 0.01 to 2 %, for example from 0.01 to 1 %, based on the weight of the organic material to be stabilised.

In addition to components (b) and (c), the compositions according to the invention may comprise further co-stabilisers (additives), such as, for example, the following:

1. Antioxidants

1.1. Alkylated monophenols, for example 2,6-di-tert-butyl-4-methylphenol, 2-tert-butyl-4,6-dimethylphenol, 2,6-di-tert-butyl-4-ethylphenol, 2,6-di-tert-butyl-4-n-butylphenol, 2,6-di-tert-butyl-4-isobutylphenol, 2,6-dicyclopentyl-4-methylphenol, 2-(α -methylcyclohexyl)-4,6-di-

methylphenol, 2,6-dioctadecyl-4-methylphenol, 2,4,6-tricyclohexylphenol, 2,6-di-tert-butyl-4-methoxymethylphenol, nonylphenols which are linear or branched in the side chains, for example, 2,6-di-nonyl-4-methylphenol, 2,4-dimethyl-6-(1'-methylundec-1'-yl)phenol, 2,4-dimethyl-6-(1'-methylheptadec-1'-yl)phenol, 2,4-dimethyl-6-(1'-methyltridec-1'-yl)phenol and mixtures thereof.

1.2. Alkylthiomethylphenols, for example 2,4-dioctylthiomethyl-6-tert-butylphenol, 2,4-dioctylthiomethyl-6-methylphenol, 2,4-dioctylthiomethyl-6-ethylphenol, 2,6-di-dodecylthiomethyl-4-nonylphenol.

1.3. Hydroquinones and alkylated hydroquinones, for example 2,6-di-tert-butyl-4-methoxyphenol, 2,5-di-tert-butylhydroquinone, 2,5-di-tert-amylhydroquinone, 2,6-diphenyl-4-octadecyloxyphenol, 2,6-di-tert-butylhydroquinone, 2,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyphenyl stearate, bis-(3,5-di-tert-butyl-4-hydroxyphenyl) adipate.

1.4. Tocopherols, for example α -tocopherol, β -tocopherol, γ -tocopherol, δ -tocopherol and mixtures thereof (Vitamin E).

1.5. Hydroxylated thiodiphenyl ethers, for example 2,2'-thiobis(6-tert-butyl-4-methylphenol), 2,2'-thiobis(4-octylphenol), 4,4'-thiobis(6-tert-butyl-3-methylphenol), 4,4'-thiobis(6-tert-butyl-2-methylphenol), 4,4'-thiobis-(3,6-di-sec-amylphenol), 4,4'-bis(2,6-dimethyl-4-hydroxyphenyl)disulfide.

1.6. Alkylidenebisphenols, for example 2,2'-methylenebis(6-tert-butyl-4-methylphenol), 2,2'-methylenebis(6-tert-butyl-4-ethylphenol), 2,2'-methylenebis[4-methyl-6-(α -methylcyclohexyl)phenol], 2,2'-methylenebis(4-methyl-6-cyclohexylphenol), 2,2'-methylenebis(6-nonyl-4-methylphenol), 2,2'-methylenebis(4,6-di-tert-butylphenol), 2,2'-ethylidenebis(4,6-di-tert-butylphenol), 2,2'-ethylidenebis(6-tert-butyl-4-isobutylphenol), 2,2'-methylenebis[6-(α -methylbenzyl)-4-nonylphenol], 2,2'-methylenebis[6-(α,α -dimethylbenzyl)-4-nonylphenol], 4,4'-methylenebis(2,6-di-tert-butylphenol), 4,4'-methylenebis(6-tert-butyl-2-methylphenol), 1,1-bis(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 2,6-bis(3-tert-butyl-5-methyl-2-hydroxybenzyl)-4-methylphenol, 1,1,3-tris(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 1,1-bis(5-tert-butyl-4-hydroxy-2-methylphenyl)-3-n-dodecylmercaptobutane, ethylene glycol bis[3,3-

bis(3'-tert-butyl-4'-hydroxyphenyl)butyrate], bis(3-tert-butyl-4-hydroxy-5-methyl-phenyl)dicyclopentadiene, bis[2-(3'-tert-butyl-2'-hydroxy-5'-methylbenzyl)-6-tert-butyl-4-methylphenyl]terephthalate, 1,1-bis-(3,5-dimethyl-2-hydroxyphenyl)butane, 2,2-bis-(3,5-di-tert-butyl-4-hydroxyphenyl)propane, 2,2-bis-(5-tert-butyl-4-hydroxy-2-methylphenyl)-4-n-dodecylmercaptobutane, 1,1,5,5-tetra-(5-tert-butyl-4-hydroxy-2-methylphenyl)pentane.

1.7. O-, N- and S-benzyl compounds, for example 3,5,3',5'-tetra-tert-butyl-4,4'-dihydroxydibenzyl ether, octadecyl-4-hydroxy-3,5-dimethylbenzylmercaptoacetate, tridecyl-4-hydroxy-3,5-di-tert-butylbenzylmercaptoacetate, tris(3,5-di-tert-butyl-4-hydroxybenzyl)amine, bis(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)dithioterephthalate, bis(3,5-di-tert-butyl-4-hydroxybenzyl)sulfide, isooctyl-3,5-di-tert-butyl-4-hydroxybenzylmercaptoacetate.

1.8. Hydroxybenzylated malonates, for example dioctadecyl-2,2-bis-(3,5-di-tert-butyl-2-hydroxybenzyl)-malonate, di-octadecyl-2-(3-tert-butyl-4-hydroxy-5-methylbenzyl)-malonate, didodecylmercaptoethyl-2,2-bis-(3,5-di-tert-butyl-4-hydroxybenzyl)malonate, bis[4-(1,1,3,3-tetramethylbutyl)phenyl]-2,2-bis(3,5-di-tert-butyl-4-hydroxybenzyl)malonate.

1.9. Aromatic hydroxybenzyl compounds, for example 1,3,5-tris-(3,5-di-tert-butyl-4-hydroxybenzyl)-2,4,6-trimethylbenzene, 1,4-bis(3,5-di-tert-butyl-4-hydroxybenzyl)-2,3,5,6-tetramethylbenzene, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxybenzyl)phenol.

1.10. Triazine Compounds, for example 2,4-bis(octylmercapto)-6-(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,3,5-triazine, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,2,3-triazine, 1,3,5-tris-(3,5-di-tert-butyl-4-hydroxybenzyl)isocyanurate, 1,3,5-tris(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)isocyanurate, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxyphenylethyl)-1,3,5-triazine, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)-hexahydro-1,3,5-triazine, 1,3,5-tris(3,5-dicyclohexyl-4-hydroxybenzyl)isocyanurate.

1.11. Benzylphosphonates, for example dimethyl-2,5-di-tert-butyl-4-hydroxybenzylphosphonate, diethyl-3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl-3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl-5-tert-butyl-4-hydroxy-3-methylbenzylphosphonate, the calcium salt of the monoethyl ester of 3,5-di-tert-butyl-4-hydroxybenzylphosphonic acid.

1.12. Acylaminophenols, for example 4-hydroxylauranilide, 4-hydroxystearanilide, octyl N-(3,5-di-tert-butyl-4-hydroxyphenyl)carbamate.

1.13. Esters of β -(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl) isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.14. Esters of β -(5-tert-butyl-4-hydroxy-3-methylphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl) isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.15. Esters of β -(3,5-dicyclohexyl-4-hydroxyphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.16. Esters of 3,5-di-tert-butyl-4-hydroxyphenyl acetic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.17. Amides of β -(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid e.g. N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hexamethylenediamide, N,N'-bis(3,5-di-tert-butyl-4-hydroxy-

phenylpropionyl)trimethylenediamide, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)-hydrazide, N,N'-bis[2-(3-[3,5-di-tert-butyl-4-hydroxyphenyl]propionyloxy)ethyl]oxamide (Nau-gard®XL-1 supplied by Uniroyal).

1.18. Ascorbic acid (vitamin C)

1.19. Aminic antioxidants, for example N,N'-di-isopropyl-p-phenylenediamine, N,N'-di-sec-butyl-p-phenylenediamine, N,N'-bis(1,4-dimethylpentyl)-p-phenylenediamine, N,N'-bis(1-ethyl-3-methylpentyl)-p-phenylenediamine, N,N'-bis(1-methylheptyl)-p-phenylenediamine, N,N'-dicyclohexyl-p-phenylenediamine, N,N'-diphenyl-p-phenylenediamine, N,N'-bis(2-naphthyl)-p-phenylenediamine, N-isopropyl-N'-phenyl-p-phenylenediamine, N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine, N-(1-methylheptyl)-N'-phenyl-p-phenylenediamine, N-cyclohexyl-N'-phenyl-p-phenylenediamine, 4-(p-toluenesulfamoyl)diphenylamine, N,N'-dimethyl-N,N'-di-sec-butyl-p-phenylenediamine, diphenylamine, N-allyldiphenylamine, 4-isopropoxydiphenylamine, N-phenyl-1-naphthylamine, N-(4-tert-octylphenyl)-1-naphthylamine, N-phenyl-2-naphthylamine, octylated diphenylamine, for example p,p'-di-tert-octyldiphenylamine, 4-n-butylaminophenol, 4-butyrylaminophenol, 4-nonanoylaminophenol, 4-dodecanoylaminophenol, 4-octadecanoylaminophenol, bis(4-methoxyphenyl)amine, 2,6-di-tert-butyl-4-dimethylaminomethylphenol, 2,4'-diaminodiphenylmethane, 4,4'-diaminodiphenylmethane, N,N,N',N'-tetramethyl-4,4'-diaminodiphenylmethane, 1,2-bis[(2-methylphenyl)-amino]ethane, 1,2-bis(phenylamino)propane, (o-tolyl)biguanide, bis[4-(1',3'-dimethylbutyl)-phenyl]amine, tert-octylated N-phenyl-1-naphthylamine, a mixture of mono- and dialkylated tert-butyl/tert-octyldiphenylamines, a mixture of mono- and dialkylated nonyldiphenylamines, a mixture of mono- and dialkylated dodecyldiphenylamines, a mixture of mono- and dialkylated isopropyl/isohexyldiphenylamines, a mixture of mono- und dialkylated tert-butyl-diphenylamines, 2,3-dihydro-3,3-dimethyl-4H-1,4-benzothiazine, phenothiazine, a mixture of mono- und dialkylated tert-butyl/tert-octylphenothiazines, a mixture of mono- und dialkylated tert-octyl-phenothiazines, N-allylphenothiazin, N,N,N',N'-tetraphenyl-1,4-diaminobut-2-ene, N,N-bis(2,2,6,6-tetramethyl-piperid-4-yl)-hexamethylenediamine, bis(2,2,6,6-tetramethylpiperid-4-yl)sebacate, 2,2,6,6-tetramethylpiperidin-4-one, 2,2,6,6-tetramethylpiperidin-4-ol.

2. UV absorbers and light stabilisers

2.1. 2-(2'-Hydroxyphenyl)benzotriazoles, for example 2-(2'-hydroxy-5'-methylphenyl)-benzotriazole, 2-(3',5'-di-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(5'-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(2'-hydroxy-5'-(1,1,3,3-tetramethylbutyl)phenyl)benzotriazole, 2-(3',5'-di-tert-butyl-2'-hydroxyphenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-methylphenyl)-5-chloro-benzotriazole, 2-(3'-sec-butyl-5'-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(2'-hydroxy-4'-octyloxyphenyl)benzotriazole, 2-(3',5'-di-tert-amyl-2'-hydroxyphenyl)benzotriazole, 2-(3',5'-bis-(α,α -dimethylbenzyl)-2'-hydroxyphenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-octyloxycarbonylethyl)phenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-5'-[2-(2-ethylhexyloxy)-carbonylethyl]-2'-hydroxyphenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-methoxycarbonylethyl)phenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-methoxycarbonylethyl)phenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-octyloxycarbonylethyl)phenyl)benzotriazole, 2-(3'-tert-butyl-5'-[2-(2-ethylhexyloxy)carbonylethyl]-2'-hydroxyphenyl)benzotriazole, 2-(3'-dodecyl-2'-hydroxy-5'-methylphenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-isooctyloxycarbonylethyl)phenyl)benzotriazole, 2,2'-methylene-bis[4-(1,1,3,3-tetramethylbutyl)-6-benzotriazole-2-ylphenol]; the transesterification product of 2-[3'-tert-butyl-5'-(2-methoxycarbonylethyl)-2'-hydroxyphenyl]-2H-benzotriazole with polyethylene glycol 300; $\left[\text{R}-\text{CH}_2\text{CH}_2-\text{COO}-\text{CH}_2\text{CH}_2 \right]_2$ where R = 3'-tert-butyl-4'-hydroxy-5'-2H-benzotriazol-2-ylphenyl, 2-[2'-hydroxy-3'-(α,α -dimethylbenzyl)-5'-(1,1,3,3-tetramethylbutyl)-phenyl]benzotriazole; 2-[2'-hydroxy-3'-(1,1,3,3-tetramethylbutyl)-5'-(α,α -dimethylbenzyl)-phenyl]benzotriazole.

2.2. 2-Hydroxybenzophenones, for example the 4-hydroxy, 4-methoxy, 4-octyloxy, 4-decyloxy, 4-dodecyloxy, 4-benzyloxy, 4,2',4'-trihydroxy and 2'-hydroxy-4,4'-dimethoxy derivatives.

2.3. Esters of substituted and unsubstituted benzoic acids, as for example 4-tertbutyl-phenyl salicylate, phenyl salicylate, octylphenyl salicylate, dibenzoyl resorcinol, bis(4-tert-butyl-benzoyl) resorcinol, benzoyl resorcinol, 2,4-di-tert-butylphenyl 3,5-di-tert-butyl-4-hydroxybenzoate, hexadecyl 3,5-di-tert-butyl-4-hydroxybenzoate, octadecyl 3,5-di-tert-butyl-4-hydroxybenzoate, 2-methyl-4,6-di-tert-butylphenyl 3,5-di-tert-butyl-4-hydroxybenzoate.

2.4. Acrylates, for example ethyl α -cyano- β,β -diphenylacrylate, isooctyl α -cyano- β,β -diphenylacrylate, methyl α -carbomethoxycinnamate, methyl α -cyano- β -methyl-p-methoxy-cinna-

mate, butyl α -cyano- β -methyl-p-methoxy-cinnamate, methyl α -carbomethoxy-p-methoxycinnamate and N-(β -carbomethoxy- β -cyanovinyl)-2-methylindoline.

2.5. Nickel compounds, for example nickel complexes of 2,2'-thio-bis-[4-(1,1,3,3-tetramethylbutyl)phenol], such as the 1:1 or 1:2 complex, with or without additional ligands such as n-butylamine, triethanolamine or N-cyclohexyldiethanolamine, nickel dibutyldithiocarbamate, nickel salts of the monoalkyl esters, e.g. the methyl or ethyl ester, of 4-hydroxy-3,5-di-tert-butylbenzylphosphonic acid, nickel complexes of ketoximes, e.g. of 2-hydroxy-4-methylphenyl undecylketoxime, nickel complexes of 1-phenyl-4-lauroyl-5-hydroxypyrazole, with or without additional ligands.

2.6. Sterically hindered amines, for example bis(2,2,6,6-tetramethyl-4-piperidyl)sebacate, bis(2,2,6,6-tetramethyl-4-piperidyl)succinate, bis(1,2,2,6,6-pentamethyl-4-piperidyl)sebacate, bis(1-octyloxy-2,2,6,6-tetramethyl-4-piperidyl)sebacate, bis(1,2,2,6,6-pentamethyl-4-piperidyl) n-butyl-3,5-di-tert-butyl-4-hydroxybenzylmalonate, the condensate of 1-(2-hydroxyethyl)-2,2,6,6-tetramethyl-4-hydroxypiperidine and succinic acid, linear or cyclic condensates of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-tert-octylamino-2,6-dichloro-1,3,5-triazine, tris(2,2,6,6-tetramethyl-4-piperidyl)nitritotriacetate, tetrakis(2,2,6,6-tetramethyl-4-piperidyl)-1,2,3,4-butane-tetracarboxylate, 1,1'-(1,2-ethanediyl)-bis(3,3,5,5-tetramethylpiperazinone), 4-benzoyl-2,2,6,6-tetramethylpiperidine, 4-stearyl-2,2,6,6-tetramethylpiperidine, bis(1,2,2,6,6-pentamethylpiperidyl)-2-n-butyl-2-(2-hydroxy-3,5-di-tert-butylbenzyl)malonate, 3-n-octyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decan-2,4-dione, bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)sebacate, bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)succinate, linear or cyclic condensates of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-morpholino-2,6-dichloro-1,3,5-triazine, the condensate of 2-chloro-4,6-bis(4-n-butylamino-2,2,6,6-tetramethylpiperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino)ethane, the condensate of 2-chloro-4,6-di-(4-n-butylamino-1,2,2,6,6-pentamethylpiperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino)ethane, 8-acetyl-3-dodecyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decane-2,4-dione, 3-dodecyl-1-(2,2,6,6-tetramethyl-4-piperidyl)pyrrolidin-2,5-dione, 3-dodecyl-1-(1,2,2,6,6-pentamethyl-4-piperidyl)pyrrolidine-2,5-dione, a mixture of 4-hexadecyloxy- and 4-stearyl-2,2,6,6-tetramethylpiperidine, a condensation product of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-cyclohexylamino-2,6-dichloro-1,3,5-triazine, a condensation product of 1,2-bis(3-aminopropylamino)ethane and 2,4,6-trichloro-1,3,5-triazine as well as

4-butylamino-2,2,6,6-tetramethylpiperidine (CAS Reg. No. [136504-96-6]); N-(2,2,6,6-tetramethyl-4-piperidyl)-n-dodecylsuccinimid, N-(1,2,2,6,6-pentamethyl-4-piperidyl)-n-dodecylsuccinimid, 2-undecyl-7,7,9,9-tetramethyl-1-oxa-3,8-diaza-4-oxo-spiro[4,5]decane, a reaction product of 7,7,9,9-tetramethyl-2-cycloundecyl-1-oxa-3,8-diaza-4-oxospiro [4,5]decane und epichlorohydrin, 1,1-bis(1,2,2,6,6-pentamethyl-4-piperidyloxycarbonyl)-2-(4-methoxyphenyl)ethene, N,N'-bis-formyl-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine, diester of 4-methoxy-methylene-malonic acid with 1,2,2,6,6-pentamethyl-4-hydroxypiperidine, poly[methylpropyl-3-oxy-4-(2,2,6,6-tetramethyl-4-piperidyl)]siloxane, reaction product of maleic acid anhydride- α -olefin-copolymer with 2,2,6,6-tetramethyl-4-aminopiperidine or 1,2,2,6,6-pentamethyl-4-aminopiperidine.

2.7. Oxamides, for example 4,4'-dioctyloxyoxanilide, 2,2'-diethoxyoxanilide, 2,2'-dioctyloxy-5,5'-di-tert-butoxanilide, 2,2'-didodecyloxy-5,5'-di-tert-butoxanilide, 2-ethoxy-2'-ethyloxanilide, N,N'-bis(3-dimethylaminopropyl)oxamide, 2-ethoxy-5-tert-butyl-2'-ethoxanilide and its mixture with 2-ethoxy-2'-ethyl-5,4'-di-tert-butoxanilide, mixtures of o- and p-methoxy-disubstituted oxanilides and mixtures of o- and p-ethoxy-disubstituted oxanilides.

2.8. 2-(2-Hydroxyphenyl)-1,3,5-triazines, for example 2,4,6-tris(2-hydroxy-4-octyloxyphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2,4-dihydroxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2,4-bis(2-hydroxy-4-propyloxyphenyl)-6-(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(4-methylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-dodecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-tridecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-butyloxy-propoxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-octyloxy-propyloxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine, 2-[4-(dodecyloxy/tridecyloxy-2-hydroxypropoxy)-2-hydroxy-phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-dodecyloxy-propoxy)phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-hexyloxy)phenyl-4,6-diphenyl-1,3,5-triazine, 2-(2-hydroxy-4-methoxyphenyl)-4,6-diphenyl-1,3,5-triazine, 2,4,6-tris[2-hydroxy-4-(3-butoxy-2-hydroxy-propoxy)phenyl]-1,3,5-triazine, 2-(2-hydroxyphenyl)-4-(4-methoxyphenyl)-6-phenyl-1,3,5-triazine, 2-[2-hydroxy-4-[3-(2-ethylhexyl-1-oxy)-2-hydroxy-propyloxy]phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine.

3. Metal deactivators, for example N,N'-diphenyloxamide, N-salicylal-N'-salicyloyl hydrazine, N,N'-bis(salicyloyl) hydrazine, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl) hydrazine, 3-salicyloylamino-1,2,4-triazole, bis(benzylidene)oxalyl dihydrazide, oxanilide, isophthaloyl dihydrazide, sebacoyl bisphenylhydrazide, N,N'-diacetyladiopoyl dihydrazide, N,N'-bis(salicyloyl)oxalyl dihydrazide, N,N'-bis(salicyloyl)thiopropionyl dihydrazide.

4. Phosphites and phosphonites, for example triphenyl phosphite, diphenyl alkyl phosphites, phenyl dialkyl phosphites, tris(nonylphenyl) phosphite, trilauryl phosphite, trioctadecyl phosphite, distearyl pentaerythritol diphosphite, tris(2,4-di-tert-butylphenyl) phosphite, diisodecyl pentaerythritol diphosphite, bis(2,4-di-tert-butylphenyl) pentaerythritol diphosphite, bis(2,6-di-tert-butyl-4-methylphenyl)-pentaerythritol diphosphite, diisodecyloxypentaerythritol diphosphite, bis(2,4-di-tert-butyl-6-methylphenyl)pentaerythritol diphosphite, bis(2,4,6-tris(tert-butylphenyl)pentaerythritol diphosphite, tristearyl sorbitol triphosphite, tetrakis(2,4-di-tert-butylphenyl) 4,4'-biphenylene diphosphonite, 6-isooctyloxy-2,4,8,10-tetra-tert-butyl-12H-dibenz[d,g]-1,3,2-dioxaphosphocin, bis(2,4-di-tert-butyl-6-methylphenyl) methyl phosphite, bis(2,4-di-tert-butyl-6-methylphenyl) ethyl phosphite, 6-fluoro-2,4,8,10-tetra-tert-butyl-12-methyl-dibenz[d,g]-1,3,2-dioxaphosphocin, 2,2',2''-nitrilo[triethyltris(3,3',5,5'-tetra-tert-butyl-1,1'-biphenyl-2,2'-diyl)phosphite], 2-ethylhexyl(3,3',5,5'-tetra-tert-butyl-1,1'-biphenyl-2,2'-diyl)phosphite, 5-butyl-5-ethyl-2-(2,4,6-tri-tert-butylphenoxy)-1,3,2-dioxaphosphirane.

5. Hydroxylamines, for example, N,N-dibenzylhydroxylamine, N,N-diethylhydroxylamine, N,N-dioctylhydroxylamine, N,N-dilaurylhydroxylamine, N,N-ditetradecylhydroxylamine, N,N-dihexadecylhydroxylamine, N,N-dioctadecylhydroxylamine, N-hexadecyl-N-octadecylhydroxylamine, N-heptadecyl-N-octadecylhydroxylamine, N,N-dialkylhydroxylamine derived from hydrogenated tallow amine.

6. Nitrones, for example, N-benzyl-alpha-phenyl-nitrone, N-ethyl-alpha-methyl-nitrone, N-octyl-alpha-heptyl-nitrone, N-lauryl-alpha-undecyl-nitrone, N-tetradecyl-alpha-tridcyl-nitrone, N-hexadecyl-alpha-pentadecyl-nitrone, N-octadecyl-alpha-heptadecyl-nitrone, N-hexadecyl-alpha-heptadecyl-nitrone, N-ocatadecyl-alpha-pentadecyl-nitrone, N-heptadecyl-alpha-heptadecyl-nitrone, N-octadecyl-alpha-hexadecyl-nitrone, nitrone derived from N,N-dialkylhydroxylamine derived from hydrogenated tallow amine.

7. Thiosynergists, for example, dilauryl thiodipropionate or distearyl thiodipropionate.
8. Peroxide scavengers, for example esters of β -thiodipropionic acid, for example the lauryl, stearyl, myristyl or tridecyl esters, mercaptobenzimidazole or the zinc salt of 2-mercaptobenzimidazole, zinc dibutyldithiocarbamate, dioctadecyl disulfide, pentaerythritol tetrakis(β -dodecylmercapto)propionate.
9. Basic co-stabilisers, for example, melamine, polyvinylpyrrolidone, dicyandiamide, triallyl cyanurate, urea derivatives, hydrazine derivatives, amines, polyamides, polyurethanes, alkali metal salts and alkaline earth metal salts of higher fatty acids for example calcium stearate, zinc stearate, magnesium behenate, magnesium stearate, sodium ricinoleate and potassium palmitate, antimony pyrocatecholate or zink pyrocatecholate.
10. Nucleating agents, for example, inorganic substances such as talcum, metal oxides such as titanium dioxide or magnesium oxide, phosphates, carbonates or sulfates of, preferably, alkaline earth metals; organic compounds such as mono- or polycarboxylic acids and the salts thereof, e.g. 4-tert-butylbenzoic acid, adipic acid, diphenylacetic acid, sodium succinate or sodium benzoate; polymeric compounds such as ionic copolymers (ionomers). Especially preferred are 1,3:2,4-bis(3',4'-dimethylbenzylidene)sorbitol, 1,3:2,4-di(parame-thyldibenzylidene)sorbitol, und 1,3:2,4-di(benzylidene)sorbitol.
11. Fillers and reinforcing agents, for example, calcium carbonate, silicates, glass fibres, glass bulbs, asbestos, talc, kaolin, mica, barium sulfate, metal oxides and hydroxides, carbon black, graphite, wood flour and flours or fibers of other natural products, synthetic fibers.
12. Triarylphosphines, for example triphenylphosphine.
13. Other additives, for example, plasticisers, lubricants, emulsifiers, pigments, rheology additives, catalysts, flow-control agents, optical brighteners, flameproofing agents, antistatic agents and blowing agents.

The co-stabilisers are added, for example, in concentrations of from 0.01 to 10 %, based on the total weight of the organic material to be stabilised.

The fillers and reinforcing agents (point 11 of the list), such as, for example, talc, calcium carbonate, mica or kaolin, are added to the polyolefins, for example, in concentrations of from 0.01 to 40 %, based on the total weight of the polyolefins to be stabilised.

The fillers and reinforcing agents (point 11 of the list), such as, for example, metal hydroxides, especially aluminium hydroxide or magnesium hydroxide, are added to the polyolefins, for example, in concentrations of from 0.01 to 60 %, based on the total weight of the polyolefins to be stabilised.

Carbon black, as filler, is added to the polyolefins advantageously in concentrations of from 0.01 to 5 %, based on the total weight of the polyolefins to be stabilised.

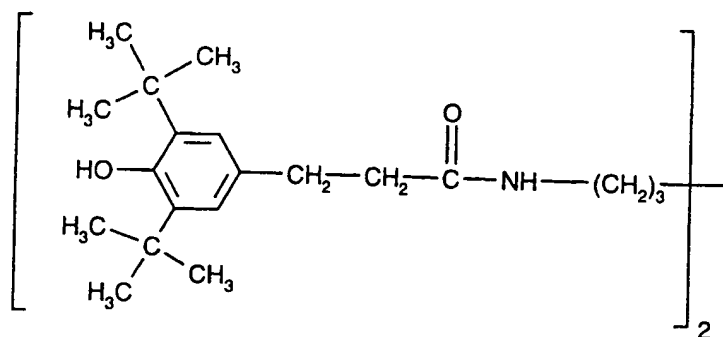
Glass fibres, as reinforcing agents, are added to the polyolefins advantageously in concentrations of from 0.01 to 20 %, based on the total weight of the polyolefins to be stabilised.

Further preferred compositions also comprise, besides components (a) to (c), further additives, especially phenolic antioxidants, light-stabilisers or processing stabilisers.

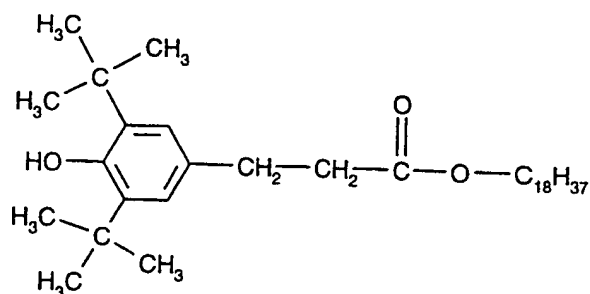
Also of interest as yet further additives in the compositions according to the invention are alkaline earth metal salts of higher fatty acids, such as, for example, calcium stearate.

Especially preferred additives are phenolic antioxidants (point 1.1 to 1.17 of the list) and peroxide-destroying compounds (point 8 of the list).

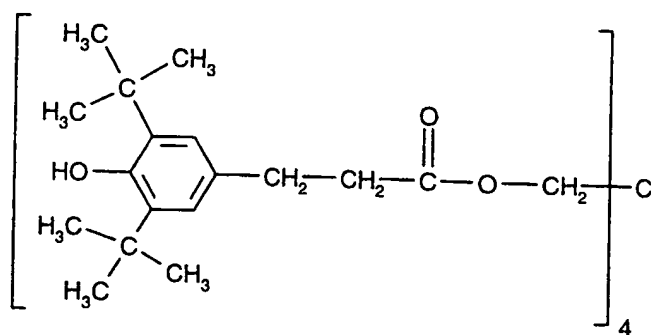
Especially preferred phenolic antioxidants are:



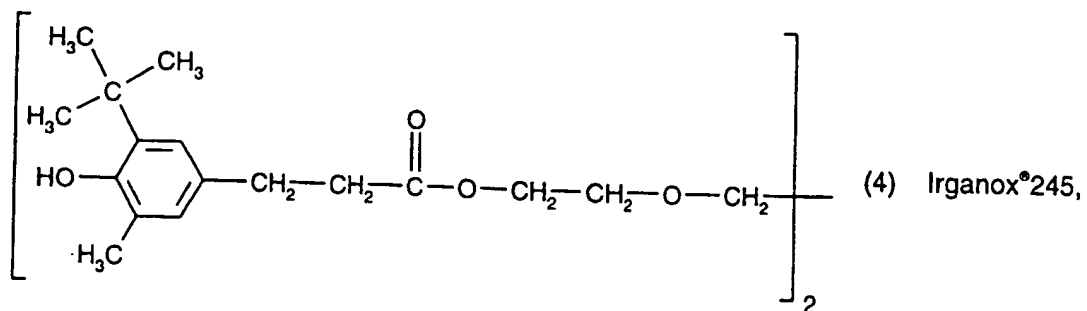
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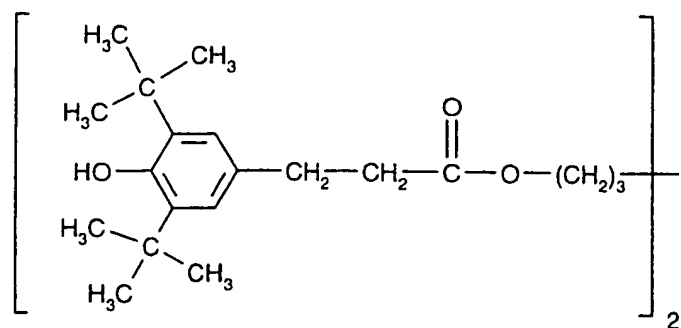
(2) Irganox®1076,



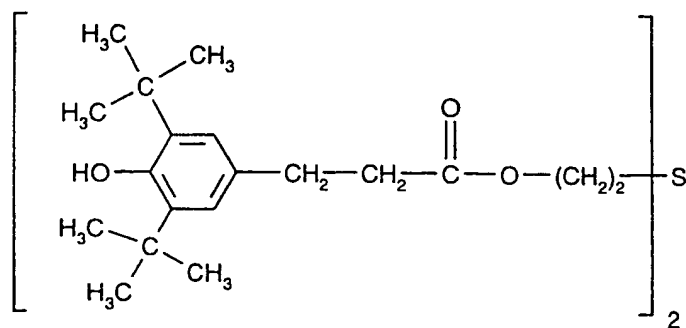
(3) Irganox®1010,



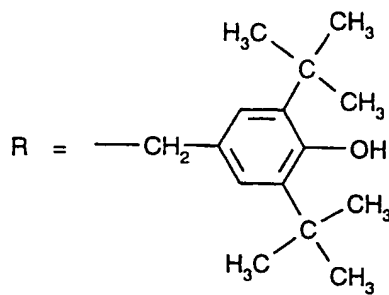
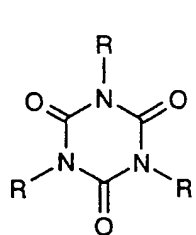
(4) Irganox®245,



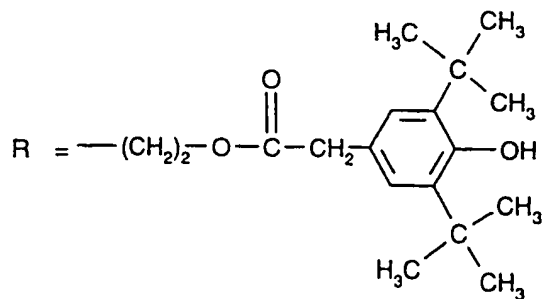
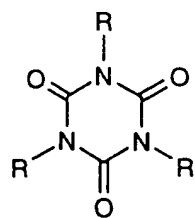
(5) Irganox®259,



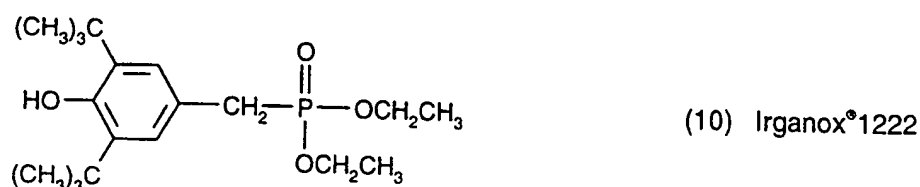
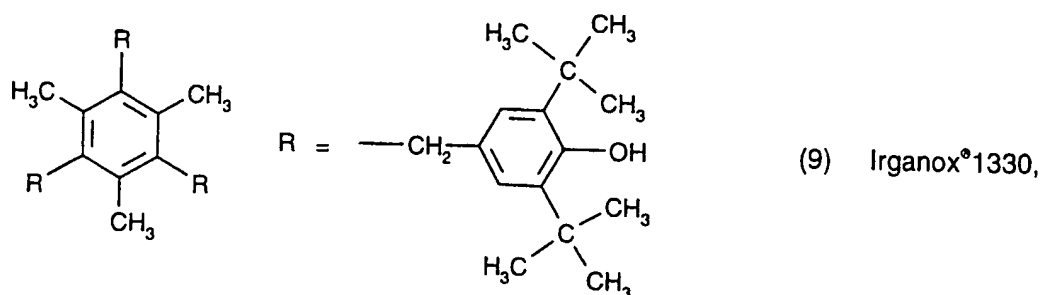
(6) Irganox®1035,



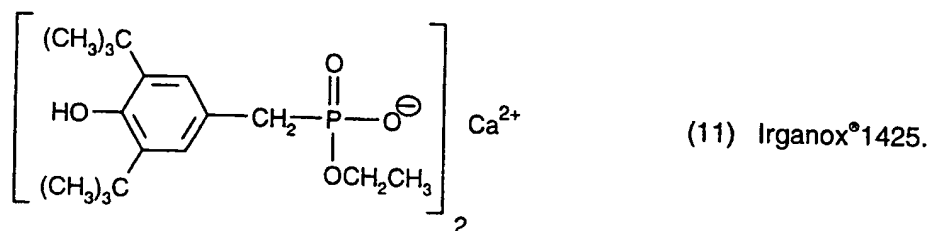
(7) Irganox®3114,



(8) Irganox®3125,



and



Irganox®1098, Irganox®1076, Irganox®1010, Irganox®245, Irganox®259, Irganox®1035, Irganox®3114, Irganox®3125, Irganox®1330, Irganox®1222 and Irganox®1425 are protected tradenames of Ciba Spezialitätenchemie AG.

As a conventional stabiliser combination for the processing of polymeric organic materials, such as, for example, polyolefins, into corresponding moulded articles, the combination of a phenolic antioxidant with a secondary antioxidant based on an organic phosphite or phosphonite is recommended. Depending on the substrate and process, however, many polyolefin processors are obliged to operate processes in the high-temperature range above approx. 280°C. Thanks to the inclusion of a processing stabiliser mixture of components (b) and (c) according to the invention, which processing stabiliser mixture is particularly suitable for high-temperature applications, especially in the temperature range above 300°C, technical materials and moulded articles for instance based on HD polyethylene, such as,

for example, pipes and their technical variants (fittings), can be manufactured with a higher output and fewer rejects. A further advantage of that stabiliser mixture is also that it can be used in a very small amount, which results in a reduction in the overall antioxidant concentration compared with conventional stabiliser mixtures. For instance the use of a low concentration of a stabiliser of the benzofuran-2-one type [component (c) (i)] allows the overall stabiliser concentration to be reduced by approximately a third in, for example, polyolefins, which at the same time represents an economic advantage.

Components (b) and (c), and also where applicable further additives, are incorporated into the polyolefin according to known methods, for example before or during the shaping, or alternatively by applying a solution or dispersion of the stabiliser mixture to the polyolefin, if necessary with subsequent evaporation of the solvent. The stabiliser mixture of components (b) and (c), and where applicable further additives, may also be added to the materials to be stabilised in the form of a master batch that contains the mixture in a concentration of, for example, from 2.5 to 25 % by weight.

The stabiliser mixture of components (b) and (c), and where applicable further additives, may also be added before or during polymerisation or before crosslinking.

The stabiliser mixture of components (b) and (c), and where applicable further additives, may be incorporated into the polyolefin to be stabilised in pure form or encapsulated in waxes, oils or polymers.

The stabiliser mixture of components (b) and (c), and where applicable further additives, may also be sprayed onto the polyolefin to be stabilised. The mixture can be used to dilute other additives (e.g. the above-mentioned conventional additives) or melts thereof, so that it is also possible for the mixture to be sprayed together with those additives onto the polymer to be stabilised. Addition by spraying during the deactivation of the polymerisation catalysts is especially advantageous, it being possible, for example, for the vapour used for the deactivation to be utilised for the spraying.

For example, where the polyolefins are polymerised in the form of beads, it may be advantageous for the stabiliser mixture of components (b) and (c), and where applicable other additives, to be applied by spraying.

The materials stabilised in that manner may be used in an extremely wide variety of forms, e.g. in the form of films, fibres, tapes, moulding compounds or profiles, or as binders for surface-coatings, especially powder coatings, adhesives or cements.

The polyolefins stabilised in that manner may likewise be used in an extremely wide variety of forms, especially in the form of thick-layer polyolefin moulded articles that are in lasting contact with extracting media, such as, for example, pipes for liquids or gases, films, fibres, geomembranes, tapes, profiles or tanks.

The preferred thick-layer polyolefin moulded articles have a layer thickness of from 1 to 50 mm, especially from 1 to 30 mm, e.g. from 2 to 10 mm.

The invention relates also to compositions comprising a functional fluid, preferably from the series of lubricants, hydraulic fluids and metal-working fluids and also fuels for powering engines of the 4-stroke, Otto, 2-stroke, diesel, Wankel and orbital types, and at least one each of components (b) and (c).

Components (b) and (c) may preferably be used in lubricants and fuels as multi-functional stabilisers, that is to say they combine in themselves antioxidative, friction-reducing, extreme-pressure-protection and wear-protection action and also anti-corrosion properties.

Preferred lubricants and fuels and related products are engine oils, turbine oils, gear oils, hydraulic fluids, diesel or Otto fuels, metal-working fluids and lubricating greases.

Especially preferred lubricants are mineral oils, synthetic oils or mixtures thereof.

Products known *per se* are used as functional fluids from the series of lubricants, hydraulic fluids and metal-working fluids.

The lubricants and hydraulic fluids that come into consideration will be familiar to the person skilled in the art and are described in the relevant specialist literature, such as, for example, in Dieter Klamann, "Schmierstoffe und verwandte Produkte" [Lubricants and related products] (Verlag Chemie, Weinheim, 1982), in Schewe-Kobek, "Das Schmiermittel-Taschen-

buch" [The lubricant handbook] (Dr. Alfred Hüthig-Verlag, Heidelberg, 1974) and in "Ullmanns Enzyklopädie der technischen Chemie" [Ullmann's Encyclopaedia of Industrial Chemistry], Vol. 13, pages 85-94 (Verlag Chemie, Weinheim, 1977).

The lubricants are especially oils and greases, for example based on a mineral oil. Oils are preferred.

A further group of lubricants that may be used are vegetable or animal oils, greases, tallows and waxes or mixtures thereof with one another or mixtures with the mentioned mineral or synthetic oils.

Vegetable and animal oils, greases, tallows and waxes are, for example, palm-kernel oil, palm oil, olive oil, rapeseed oil, rape oil, linseed oil, groundnut oil, soybean oil, cottonseed oil, sunflower oil, pumpkin seed oil, coconut oil, maize oil, castor oil, tree nut oil and mixtures thereof, fish oils, tallows obtained from slaughtered animals, such as beef tallow, neatsfoot oil and bone oil, and modified, epoxidised and sulfoxidised forms thereof, for example epoxidised soybean oil.

The mineral oils are based especially on hydrocarbon compounds.

Examples of synthetic lubricants include lubricants based on aliphatic or aromatic carboxy esters, polymeric esters, polyalkylene oxides, phosphoric acid esters, poly-alpha-olefins or silicones, a diester of a divalent acid with a monohydric alcohol, such as, for example, dioctyl sebacate or dinonyl adipate, a triester of trimethylolpropane with a monovalent acid or with a mixture of such acids, such as, for example, trimethylolpropane tripelargonate, trimethylolpropane tricaprylate or mixtures thereof, a tetraester of pentaerythritol with a monovalent acid or with a mixture of such acids, such as, for example, pentaerythritol tetracaprylate, or a complex ester of monovalent and divalent acids with polyhydric alcohols, for example a complex ester of trimethylolpropane with caprylic and sebacic acid, or a mixture thereof. Apart from mineral oils there are especially suitable, for example, poly-alpha-olefins, ester-based lubricants, phosphates, glycols, polyglycols and polyalkylene glycols, and also mixtures thereof with water.

Metal-working fluids and hydraulic fluids may be prepared on the basis of the same substances as those described above for the lubricants, such fluids frequently being emulsions of such substances in water or other liquids.

Lubricant and fuel compositions according to the invention are used, for example, in internal combustion engines, e.g. in motorised vehicles equipped with, for example, engines of the Otto, diesel, two-stroke, Wankel or orbital type.

Components (b) and (c) are readily soluble in lubricants and fuels, metal-working fluids and hydraulic fluids and are therefore especially suitable as additives for lubricants and fuels, metal-working fluids and hydraulic fluids.

As additives in lubricants, components (b) and (c) are effective even in very small amounts. They are mixed in with the lubricants advantageously in an amount of from 0.01 to 5 % by weight, preferably in an amount of from 0.05 to 3 % by weight and very especially in an amount of from 0.1 to 2 % by weight, in each case based on the lubricant.

Components (b) and (c) may be mixed in with the lubricants and fuels in a manner known *per se*. Components (b) and (c) are readily soluble, for example, in oils. It is also possible to prepare a so-called master batch, which may be diluted, as a function of use, with the appropriate lubricant or fuel to the concentrations suitable for use. In such cases concentrations above 1 % by weight are possible.

The lubricants and fuels, metal-working fluids and hydraulic fluids may additionally comprise other additives that are added in order to improve their basic properties still further; such additives include: further antioxidants, metal passivators, rust inhibitors, viscosity index improvers, pour-point depressants, dispersants, detergents, coefficient of friction reducers, further extreme-pressure additives and anti-wear additives. Such further additives are added advantageously in an amount of from 0.01 to 5 % by weight.

A number of such compounds can be found, for example, in the above list "1. Antioxidants", especially points 1.1 to 1.19. In addition, further additives may be mentioned by way of example:

Examples of further antioxidants:

Aliphatic or aromatic phosphites, esters of thiodipropionic acid or thiodiacetic acid or salts of dithiocarbamic or dithiophosphoric acid, 2,2,12,12-tetramethyl-5,9-dihydroxy-3,7,11-trithiatridecane and 2,2,15,15-tetramethyl-5,12-dihydroxy-3,7,10,14-tetrathiahexadecane.

Examples of metal deactivators, e.g. for copper, are:

- a) Benzotriazoles and derivatives thereof, e.g. 2-mercaptobenzotriazole, 2,5-dimercaptobenzotriazole, 4- or 5-alkylbenzotriazoles (e.g. tolutriazole) and derivatives thereof, 4,5,6,7-tetrahydrobenzotriazole, 5,5'-methylenebis-benzotriazole; Mannich bases of benzotriazole or tolutriazole, such as 1-[di(2-ethylhexyl)aminomethyl]tolutriazole and 1-[di(2-ethylhexyl)aminomethyl]benzotriazole; alkoxyalkylbenzotriazoles, such as 1-(nonyloxymethyl)benzotriazole, 1-(1-butoxyethyl)benzotriazole and 1-(1-cyclohexyloxybutyl)tolutriazole.
- b) 1,2,4-Triazoles and derivatives thereof, e.g. 3-alkyl- (or -aryl-)1,2,4-triazoles, Mannich bases of 1,2,4-triazoles, such as 1-[di(2-ethylhexyl)aminomethyl]-1,2,4-triazole; alkoxyalkyl-1,2,4-triazoles, such as 1-(1-butoxyethyl)-1,2,4-triazole; acylated 3-amino-1,2,4-triazoles.
- c) Imidazole derivatives, e.g. 4,4'-methylenebis(2-undecyl-5-methyl)imidazole and bis[(N-methyl)imidazol-2-yl]carbinol-octyl ether.
- d) Sulfur-containing heterocyclic compounds, e.g. 2-mercaptobenzothiazole, 2,5-dimercapto-1,3,4-thiadiazole, 2,5-dimercaptobenzothiadiazole and derivatives thereof; 3,5-bis[di(2-ethylhexyl)aminomethyl]-1,3,4-thiadiazolin-2-one.
- e) Amino compounds, e.g. salicylidene-propylenediamine, salicylaminoguanidine and salts thereof.

Examples of rust inhibitors are:

- a) Organic acids, their esters, metal salts, amine salts and anhydrides, e.g. alkyl- and alkenyl-succinic acids and their partial esters with alcohols, diols or hydroxycarboxylic acids, partial amides of alkyl- and alkenyl-succinic acids, 4-nonylphenoxyacetic acid,

alkoxy- and alkoxyethoxy-carboxylic acids, such as dodecyloxyacetic acid, dodecyloxy-(ethoxy)acetic acid and amine salts thereof, and also N-oleoyl-sarcosine, sorbitan monooleate, lead naphthenate, alkenylsuccinic acid anhydrides, e.g. dodecenylsuccinic acid anhydride, 2-(2-carboxyethyl)-1-dodecyl-3-methylglycerol and salts thereof, especially sodium and triethanolamine salts thereof.

- b) Nitrogen-containing compounds, e.g.:
 - i. Primary, secondary or tertiary, aliphatic or cycloaliphatic amines and amine salts of organic and inorganic acids, e.g. oil-soluble alkylammonium carboxylates, and 1-[N,N-bis(2-hydroxyethyl)amino]-3-(4-nonylphenoxy)propan-2-ol.
 - ii. Heterocyclic compounds, e.g.: substituted imidazolines and oxazolines, e.g. 2-heptadecenyl-1-(2-hydroxyethyl)-imidazoline.
- c) Phosphorus-containing compounds, e.g.:

Amine salts of phosphoric acid partial esters or phosphonic acid partial esters, zinc dialkyldithiophosphates.
- d) Sulfur-containing compounds, e.g.:

Barium dinonylnaphthalene sulfonates, calcium petroleum sulfonates, alkylthio-substituted aliphatic carboxylic acids, esters of aliphatic 2-sulfocarboxylic acids and salts thereof.
- e) Glycerol derivatives, e.g.:

Glycerol monooleate, 1-(alkylphenoxy)-3-(2-hydroxyethyl)glycerols, 1-(alkylphenoxy)-3-(2,3-dihydroxypropyl)glycerols, 2-carboxyalkyl-1,3-dialkylglycerols.

Examples of viscosity index improvers are:

Polyacrylates, polymethacrylates, vinylpyrrolidone/methacrylate copolymers, polyvinylpyrrolidones, polybutenes, olefin copolymers, styrene/acrylate copolymers, polyethers.

Examples of pour-point depressants are:

Poly(meth)acrylates, ethylene/vinyl acetate copolymer, alkylpolystyrenes, fumarate copolymers, alkylated naphthalene derivatives.

Examples of dispersants/surfactants are:

Polybutenylsuccinic acid amides or imides, polybutenylphosphonic acid derivatives, basic magnesium, calcium and barium sulfonates and phenolates.

Examples of extreme-pressure and anti-wear additives are:

Sulfur- and/or phosphorus- and/or halogen-containing compounds, such as, for example, chlorinated paraffins, sulfurated olefins or vegetable oils (soybean/rape oil), alkyl- or aryl-di- or -tri-sulfides, zinc dialkyldithiophosphates, zinc dithiocarbamates such as zinc diamyldithiocarbamate, molybdenum dithioates such as molybdenum dithiocarbamates, triaryl phosphates such as tritolyl phosphate, tricresyl phosphate, phenyl phosphate isopropyl ester, amine salts of mono- or di-alkylphosphoric acids such as the amine salts of mono-/di-hexyl phosphate, amine salts of alkylphosphonic acids such as the amine salt of methylphosphonic acid, triaryl phosphites such as tris[nonylphenyl] phosphite, dialkyl phosphites such as dioctyl phosphite, triaryl monothiophosphates such as triphenyl thionophosphate or tris[isnonylphenyl] thionophosphate or tert-butylated triphenyl thionophosphate, substituted tri-alkyl mono- or di-thiophosphates such as diisopropoxyphosphinothioyl[thio]propionate or butylene-1,3-bis[(diisobutoxyphosphinothioyl)propionate, trithiophosphates such as trithiophosphoric acid S,S,S-tris(isooctyl-2-acetates), amine salts of 3-hydroxy-1,3-thiaphosphatane-3-oxide, benzotriazoles or derivatives thereof such as bis(2-ethylhexyl)aminomethyltolutriazole, dithiocarbamates such as methylene-bis-dibutyldithiocarbamate, derivatives of 2-mercaptobenzothiazole such as 1-[N,N-bis(2-ethylhexyl)aminomethyl]-2-mercapto-1H-1,3-benzothiazole, derivatives of 2,5-dimercapto-1,3,4-thiadiazole such as 2,5-bis(tert-nonyldithio)-1,3,4-thiadiazole.

Examples of coefficient of friction reducers are:

Lard oil, oleic acid, tallow, rape oil, sulfurated fats, amines. Further examples are given in EP-A-0 565 487.

Examples of special additives for use in water/oil metal-working fluids and hydraulic fluids are:

Emulsifiers: petroleum sulfonates, amines, such as polyoxyethylated fatty amines, non-ionic surface-active substances;

buffers: alkanolamines;

biocides: triazines, thiazolinones, tris-nitromethane, morpholine, sodium pyridenethol;

speed improvers: calcium and barium sulfonates;

Examples of fuel additives:

Fuel additives are described in Kirk-Othmer, Encyclopedia of Chemical Technology, Vol 12, 1994 and in this instance are essentially petrol and diesel additives:

Petrol: dyes, especially azo dyes;

Antioxidants: aminic, especially para-phenylenediamines, or phenolic, e.g. 2,6-di-tert-butyl-phenol, as described above;

Metal deactivators: especially N,N'-disalicylidene-1,2-propane, benzotriazole, EDTA;

Rust inhibitors: for example carboxylic acids, sulfonates, amines or amine salts;

Dispersants: e.g. esters, high-molecular-weight amines, Mannich bases, succinimides, borated succinimides;

Detergents: for example fatty acid amides, nonpolymeric amines, polybutene succinimides, polyether amines, low-molecular-weight amines, sulfonates, salicylic acid derivatives;

Demulsifiers: for example long-chain alcohols or phenols containing poly-ethylene or -butylene groups;

Antiknock agents: tetraalkyl lead, manganese methylcyclopentadienyltricarbonyl;

Oxygen compounds: esters of vegetable oils, ethers, alcohols for improving burn behaviour;

Diesel: ignition improvers (cetane improvers), e.g. alkyl nitrates, ether nitrates, alkyl diglycol nitrates, organic peroxides;

Stabilisers for, especially, cracked diesel: amines and other N-containing compounds that act as radical traps.

Especially preferred further additives in lubricants are aminic antioxidants, especially mixtures of mono- and di-alkylated tert-butyl-/tert-octyl-diphenylamines.

The present invention relates also to the use of components (b) and (c) for stabilising organic materials, especially as additives in lubricants and fuels, hydraulic fluids or metal-working fluids, preferably in hydraulic oils and gear oils. The use according to the invention includes protection of the metal components to be lubricated against mechanical attrition (wear protection) and corrosion protection activity and also antioxidation activity - with respect both to the lubricant and to the metal components.

The present invention accordingly relates also to a method for improving the properties, during use, of organic materials, especially lubricants and fuels, metal-working fluids and hydraulic fluids, wherein components (b) and (c) are added to those materials.

It is also possible to prepare concentrates comprising an oleophilic solvent and at least one each of components (b) and (c).

A preferred embodiment of the present invention is likewise the use of components (b) and (c) as stabilisers, especially processing stabilisers (thermostabilisers), for organic materials against oxidative, thermal or light-induced degradation.

The present invention relates also to a stabiliser mixture comprising (α) at least one compound of formula A and (β) (i) at least one compound of the benzofuran-2-one type or (β) (ii) α -tocopherol.

The present invention relates likewise to a stabiliser mixture comprising, in addition, (γ) at least one compound from the group of phenolic antioxidants.

Preference is given also to stabiliser mixtures wherein the weight ratio of components (α) : (β) is from 100 : 0.01 to 0.01 : 100.

Preference is given likewise to stabiliser mixtures wherein the weight ratio of components $(\alpha) : (\beta) : (\gamma)$ is from 100 : 1 : 0.01 to 0.01 : 1 : 100, especially from 5 : 1 : 0.1 to 0.01 : 1 : 10.

The stabiliser mixtures according to the invention, which comprise components (b) and (c) and where applicable further additives, are distinguished by an outstandingly good stability towards hydrolysis and an advantageous colour behaviour, that is to say little discoloration of the organic materials, especially polyolefins, during processing.

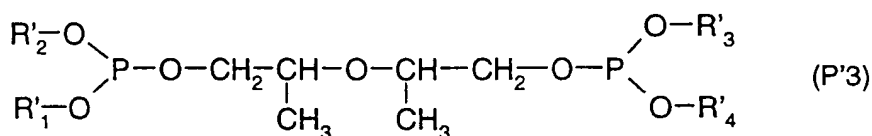
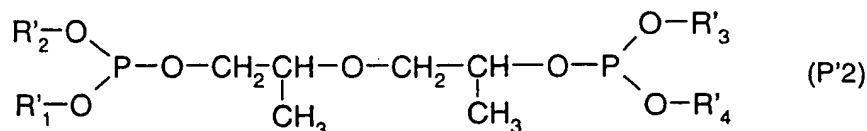
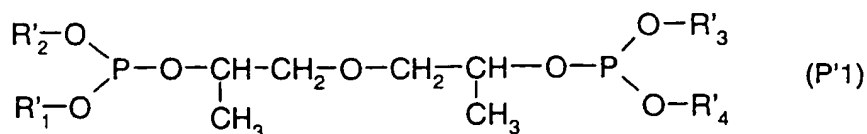
The present invention relates also to a method for stabilising organic materials against oxidative, thermal or light-induced degradation wherein at least one each of components (b) and (c) are incorporated therein or applied thereto.

The preferred components (b) and (c) for use as stabilisers, the method of stabilisation and the stabiliser mixture are the same as those described for the compositions comprising an organic material.

The following Examples illustrate the invention further. Parts or percentages relate to weight.

Example 1: Preparation of tetra-isodecyl-dipropylene glycol bisphosphite [compound (P1)].

310.3 g (1.0 mol) of triphenyl phosphite, 67.1 g (0.5 mol) of dipropylene glycol (mixture of isomers), 316.6 g (2.0 mol) of isodecanol (mixture of isomers) and 2.7 g (0.05 mol) of sodium methylate are placed in a reaction apparatus well flushed with dried nitrogen and equipped with a stirring mechanism, thermometer, distillation head and receiver flask, heated up to 110°C within one hour and stirred at 110°C for 3 hours. After cooling to 80°C, the major portion of the liberated phenol is distilled off *in vacuo* while slowly increasing the temperature to 150°C and the remainder of the phenol is drawn off within one hour at 150°C and approx. 0.1 mbar using an oil pump. 2 g of filtration aid are added to the residue and the mixture is filtered. 376 g of a light-yellow liquid [compound (P1)] having a refractive index n_D^{20} of 1.4620 are obtained. Analysis, calculated: 7.53 % P. Analysis, found: 7.59 %. The compound (P1) is a mixture of isomers of monomeric and oligomeric compounds having main components of formulae P'1, P'2 and P'3:



wherein R'₁, R'₂, R'₃ and R'₄ are $\text{CH}_3 \text{---} \underset{\text{CH}_3}{\text{CH}} \text{---} \underset{\text{CH}_3}{\text{CH}} \text{---} \underset{\text{CH}_3}{\text{CH}} \text{---} \text{CH}_2 \text{---}$.

Example 2: Stabilisation of polypropylene during multiple extrusion.

1.5 kg of polypropylene powder (Polychim®A 10 TB), which has been pre-stabilised using 0.051 % Irganox®1010 (compound of formula 3), (having a melt index of 3.2 measured at 230°C using 2.16 kg), are mixed with 0.050 % zinc stearate and the stabilisers given in Table 1. That mixture is extruded in an extruder having a cylinder diameter of 20 mm and a length of 400 mm at 100 revolutions per minute, the maximum extruder temperature being set at 280°C. The extrudate is drawn through a water bath for the purpose of cooling and is then granulated. That granulate is extruded repeatedly. After 5 extrusions, the melt index is measured (at 180°C using 2.16 kg). A large increase in the melt index indicates extensive chain degradation, that is to say poor stabilisation. The results are given in Table 1. Test plates 44 mm by 20 mm in size and 2 mm thick are pressed from the granulate of the individual formulations (Examples 2a and 2b) using a table press. The Yellowness Index (YI) of those test plates is determined according to ASTM D 1925-70. Low YI values indicate little

discoloration; high YI values indicate severe discoloration. The less discoloration there is, the more effective is the stabiliser or stabiliser mixture. The results are given in Table 1.

Table 1:

Examples	Stabilisers	Melt Index after 5 extrusions	Yellowness Index
Example 2a ^{a)}	0.051 % Irgafos®168 ^{c)} 0.018 % HP 136 ^{d)}	2.92	2.3
Example 2b ^{b)}	0.051 % compound P1 ^{e)} 0.018 % HP 136 ^{d)}	2.21	0.0

For footnotes a) to e) see the end of Table 3.

Example 3: Stabilisation of polypropylene during multiple extrusion.

1.5 kg of polypropylene powder (Polychim®A 10 TB) are mixed with 0.050 % zinc stearate and the stabilisers given in Table 2. That mixture is extruded in an extruder having a cylinder diameter of 20 mm and a length of 400 mm at 100 revolutions per minute, the maximum extruder temperature being set at 280°C. The extrudate is drawn through a water bath for the purpose of cooling and is then granulated. That granulate is extruded repeatedly. After 5 extrusions, the melt index is measured (at 180°C using 2.16 kg). A large increase in the melt index indicates extensive chain degradation, that is to say poor stabilisation. The results are given in Table 2. Test plates 44 mm by 20 mm in size and 2 mm thick are pressed from the granulate of the individual formulations (Examples 3a and 3b) using a table press. The Yellowness Index (YI) of those test plates is determined according to ASTM D 1925-70. Low YI values indicate little discoloration; high YI values indicate severe discoloration. The less discoloration there is, the more effective is the stabiliser or stabiliser mixture. The results are given in Table 2.

Table 2:

Examples	Stabilisers	Melt Index after 5 extrusions	Yellowness Index
Example 3a ^{a)}	0.03 % Irgafos®168 ^{c)} 0.02 % compound 101 ^{f)}	26.0	5.3
Example 3b ^{b)}	0.03 % compound P1 ^{e)} 0.02 % compound 101 ^{f)}	20.3	3.0

For footnotes a), b), c), e) and f) see the end of Table 3.

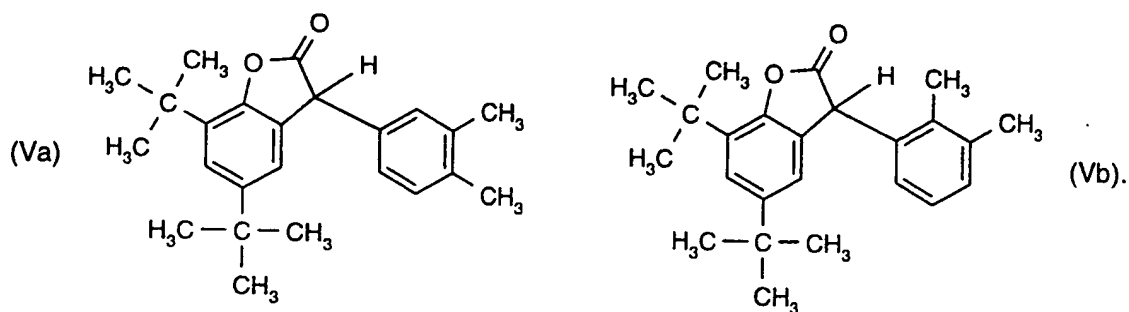
Example 4: Stabilisation of polyethylene during multiple extrusion.

1.5 kg of polyethylene powder (LLDPE Dowlex®2045 E), which has been pre-stabilised using 0.0168 % Irganox®1076 (compound of formula 2), (having a melt index of 3.2 measured at 230°C using 2.16 kg), are mixed with 0.050 % zinc stearate and the stabilisers given in Table 3. That mixture is extruded in an extruder having a cylinder diameter of 20 mm and a length of 400 mm at 100 revolutions per minute, the maximum extruder temperature being set at 280°C. The extrudate is drawn through a water bath for the purpose of cooling and is then granulated. That granulate is extruded repeatedly. After 5 extrusions, test plates 44 mm by 20 mm in size and 2 mm thick are pressed from the granulate of the individual formulations (Examples 4a and 4b) using a table press. The Yellowness Index (YI) of those test plates is determined according to ASTM D 1925-70. Low YI values indicate little discoloration; high YI values indicate severe discoloration. The less discoloration there is, the more effective is the stabiliser or stabiliser mixture. The results are given in Table 3.

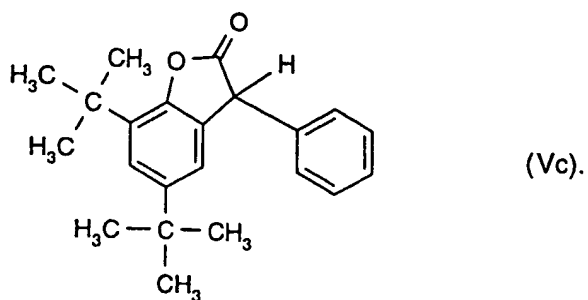
Table 3:

Examples	Stabilisers	Yellowness Index
Example 4a ^{a)}	0.068 % TNPP ^{g)} 0.0075 % compound 101 ^{f)}	25.6
Example 4b ^{b)}	0.068 % compound P1 ^{e)} 0.0075 % compound 101 ^{f)}	15.5

- a) Example for comparison.
- b) Example according to the invention.
- c) Irgafos®168 (Ciba Spezialitätenchemie AG) is tris(2,4-di-tert-butylphenyl) phosphite.
- d) HP 136 (Ciba Spezialitätenchemie AG) is a mixture of approx. 85 parts by weight of the compound of formula Va and approx. 15 parts by weight of the compound of formula Vb



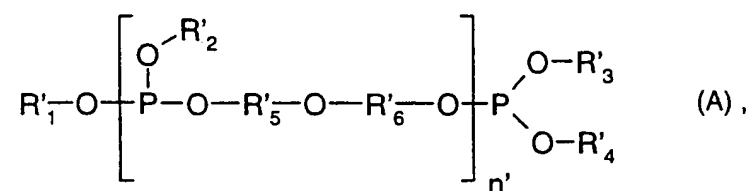
- e) The preparation of compound P1 is disclosed in Example 1.
- f) Compound 101 is a compound of formula Vc



- g) TNPP is tris(nonylphenyl) phosphite.

What is claimed is:

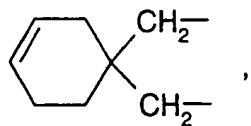
1. A composition comprising
 - a) an organic material subject to oxidative, thermal or light-induced degradation,
 - b) at least one compound of formula A



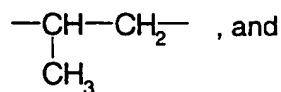
wherein

n' is a number from 1 to 6,

R'_1 , R'_2 , R'_3 and R'_4 are each independently of the others C_4 - C_{22} alkyl, C_7 - C_9 phenylalkyl; or C_5 - C_8 cycloalkyl unsubstituted or substituted by C_1 - C_4 alkyl; or R'_1 and R'_2 together and/or R'_3 and R'_4 together are C_2 - C_9 alkylene or a group of the formula



R'_5 and R'_6 are each independently of the other $-\text{CH}_2-\text{CH}_2-$ or



- c) (i) at least one compound of the benzofuran-2-one type or (ii) α -tocopherol.

2. A composition according to claim 1, comprising as component (b) a compound of formula A wherein n' is from 1 to 4.

3. A composition according to claim 1, comprising as component (b) a compound of formula A wherein

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R'₁, R'₂, R'₃ and R'₄ are each independently of the others C₆-C₂₂alkyl or C₇-C₉phenylalkyl; or R'₁ and R'₂ together and/or R'₃ and R'₄ together are C₂-C₇alkylene, and

R'₅ and R'₆ are each independently of the other $\text{—CH}_2\text{—CH}_2\text{—}$ or $\text{—}\underset{\text{CH}_3}{\text{CH}}\text{—CH}_2\text{—}$.

4. A composition according to claim 1, comprising as component (b) a compound of formula A wherein

R'₁, R'₂, R'₃ and R'₄ are each independently of the others C₆-C₁₅alkyl or C₇-C₉phenylalkyl; or R'₁ and R'₂ together and/or R'₃ and R'₄ together are C₂-C₄alkylene, and

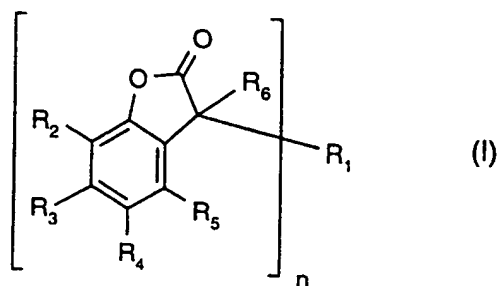
R'₅ and R'₆ each is $\text{—}\underset{\text{CH}_3}{\text{CH}}\text{—CH}_2\text{—}$.

5. A composition according to claim 1, comprising as component (b) a compound of formula A wherein

R'₁, R'₂, R'₃ and R'₄ are each independently of the others C₈-C₁₂alkyl, and

R'₅ and R'₆ each is $\text{—}\underset{\text{CH}_3}{\text{CH}}\text{—CH}_2\text{—}$.

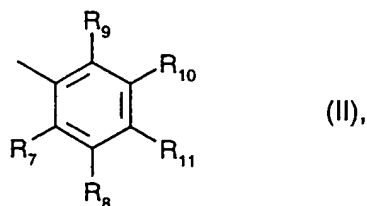
6. A composition according to claim 1, comprising as component (c) (i) a compound of formula I



wherein,

when n is 1,

R₁ is naphthyl, phenanthryl, anthryl, 5,6,7,8-tetrahydro-2-naphthyl, 5,6,7,8-tetrahydro-1-naphthyl, thienyl, benzo[b]thienyl, naphtho[2,3-b]thienyl, thianthrenyl, dibenzofuryl, chromenyl, xanthenyl, phenoxathiinyl, pyrrolyl, imidazolyl, pyrazolyl, pyrazinyl, pyrimidinyl, pyridazinyl, indoliziny, isoindolyl, indolyl, indazolyl, purinyl, quinoliziny, isoquinolyl, quinolyl, phthalazinyl, naphthyridinyl, quinoxaliny, quinazolinyl, cinnoliny, pteridinyl, carbazolyl, β-carbolinyl, phenanthridinyl, acridinyl, perimidinyl, phenanthrolinyl, phenazinyl, isothiazolyl, phenothiazinyl, isoxazolyl, furazanyl, biphenyl, terphenyl, fluorenyl or phenoxazinyl each unsubstituted or substituted by C₁-C₄alkyl, C₁-C₄alkoxy, C₁-C₄alkylthio, hydroxy, halogen, amino, C₁-C₄alkylamino, phenylamino or by di(C₁-C₄alkyl)amino, or R₁ is a radical of formula II



and,

when n is 2,

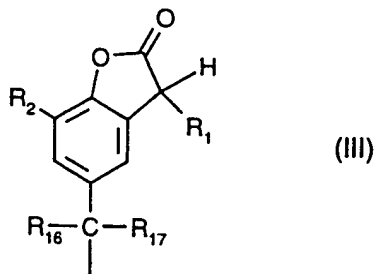
R₁ is phenylene or naphthylene each unsubstituted or substituted by C₁-C₄alkyl or by hydroxy; or -R₁₂-X-R₁₃- ,

R₂, R₃, R₄ and R₅ are each independently of the others hydrogen, chlorine, hydroxy, C₁-C₂₅-alkyl, C₇-C₉phenylalkyl; phenyl unsubstituted or substituted by C₁-C₄alkyl; C₅-C₈cycloalkyl unsubstituted or substituted by C₁-C₄alkyl; C₁-C₁₈alkoxy, C₁-C₁₈alkylthio, C₁-C₄alkylamino, di(C₁-C₄alkyl)amino, C₁-C₂₅alkanoyloxy, C₁-C₂₅alkanoylamino, C₃-C₂₅alkenoyloxy; C₃-C₂₅-

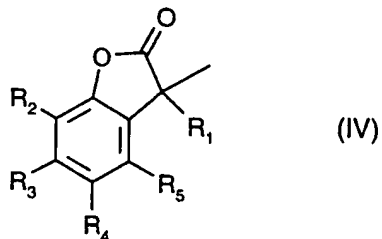
alkanoyloxy interrupted by oxygen, sulfur or by >N-R_{14} ; C₆-C₉cycloalkylcarbonyloxy,

benzoyloxy, or benzoyloxy substituted by C₁-C₁₂alkyl; or furthermore the radicals R₂ and R₃ or the radicals R₃ and R₄ or the radicals R₄ and R₅, together with the carbon atoms to which

they are bonded, form a benzo ring, in addition R_4 is $-(CH_2)_p-COR_{15}$ or $-(CH_2)_qOH$ or in addition, when R_3 , R_5 and R_6 are hydrogen, R_4 is a radical of formula III



wherein R_1 is defined as described above for when $n = 1$,
 R_6 is hydrogen or a radical of formula IV



wherein R_4 is not a radical of formula III and R_1 is defined as described above for when $n = 1$,

R_7 , R_8 , R_9 , R_{10} and R_{11} are each independently of the others hydrogen, halogen, hydroxy,

C_1 - C_{25} alkyl; C_2 - C_{25} alkyl interrupted by oxygen, sulfur or by $\text{>N}-R_{14}$; C_1 - C_{25} alkoxy;

C_2 - C_{25} alkoxy interrupted by oxygen, sulfur or by $\text{>N}-R_{14}$; C_1 - C_{25} alkylthio, C_3 - C_{25} -

alkenyl, C_3 - C_{25} alkenyloxy, C_3 - C_{25} alkynyl, C_3 - C_{25} alkynyloxy, C_7 - C_9 phenylalkyl, C_7 - C_9 phenylalkoxy; phenyl unsubstituted or substituted by C_1 - C_4 alkyl; phenoxy unsubstituted or substituted by C_1 - C_4 alkyl; C_5 - C_8 cycloalkyl unsubstituted or substituted by C_1 - C_4 alkyl; C_5 - C_8 cycloalkoxy unsubstituted or substituted by C_1 - C_4 alkyl; C_1 - C_4 alkylamino, di(C_1 - C_4 alkyl)amino,

C₁-C₂₅alkanoyl; C₃-C₂₅alkanoyl interrupted by oxygen, sulfur or by >N-R_{14} ; C₁-C₂₅-

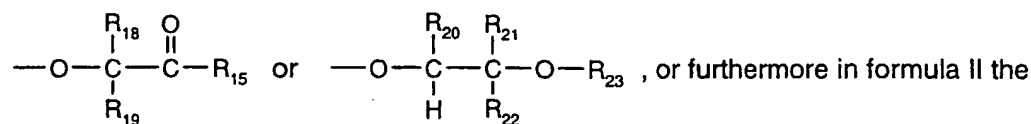
alkanoyloxy; C₃-C₂₅alkanoyloxy interrupted by oxygen, sulfur or by >N-R_{14} ; C₁-C₂₅-

alkanoylamino, C₃-C₂₅alkenoyl; C₃-C₂₅alkenoyl interrupted by oxygen, sulfur or by

>N-R_{14} ; C₃-C₂₅alkenoyloxy; C₃-C₂₅alkenoyloxy interrupted by oxygen, sulfur or by

>N-R_{14} ; C₆-C₉cycloalkylcarbonyl, C₆-C₉cycloalkylcarbonyloxy, benzoyl, or benzoyl

substituted by C₁-C₁₂alkyl; benzoyloxy, or benzoyloxy substituted by C₁-C₁₂alkyl;



radicals R₇ and R₈ or the radicals R₈ and R₁₁, together with the carbon atoms to which they are bonded, form a benzo ring,

R₁₂ and R₁₃ are each independently of the other phenylene or naphthylene each unsubstituted or substituted by C₁-C₄alkyl,

R₁₄ is hydrogen or C₁-C₈alkyl,

R₁₅ is hydroxy, $\left[-\text{O}^- \frac{1}{r} \text{M}^{r+} \right]$, C₁-C₁₈alkoxy or $\text{>N} \begin{array}{l} \text{R}_{24} \\ \text{R}_{25} \end{array}$,

R₁₆ and R₁₇ are each independently of the other hydrogen, CF₃, C₁-C₁₂alkyl or phenyl, or R₁₆ and R₁₇, together with the carbon atom to which they are bonded, form a C₅-C₈cycloalkylidene ring unsubstituted or substituted by from 1 to 3 C₁-C₄alkyl groups;

R₁₈ and R₁₉ are each independently of the other hydrogen, C₁-C₄alkyl or phenyl,

R₂₀ is hydrogen or C₁-C₄alkyl,

R₂₁ is hydrogen; phenyl unsubstituted or substituted by C₁-C₄alkyl; C₁-C₂₅alkyl; C₂-C₂₅alkyl

interrupted by oxygen, sulfur or by >N-R_{14} ; C₇-C₉phenylalkyl unsubstituted or substituted

tuted on the phenyl moiety by from 1 to 3 C₁-C₄alkyl groups; C₇-C₂₅phenylalkyl interrupted

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by oxygen, sulfur or by $\text{N}-\text{R}_{14}$ and unsubstituted or substituted on the phenyl moiety

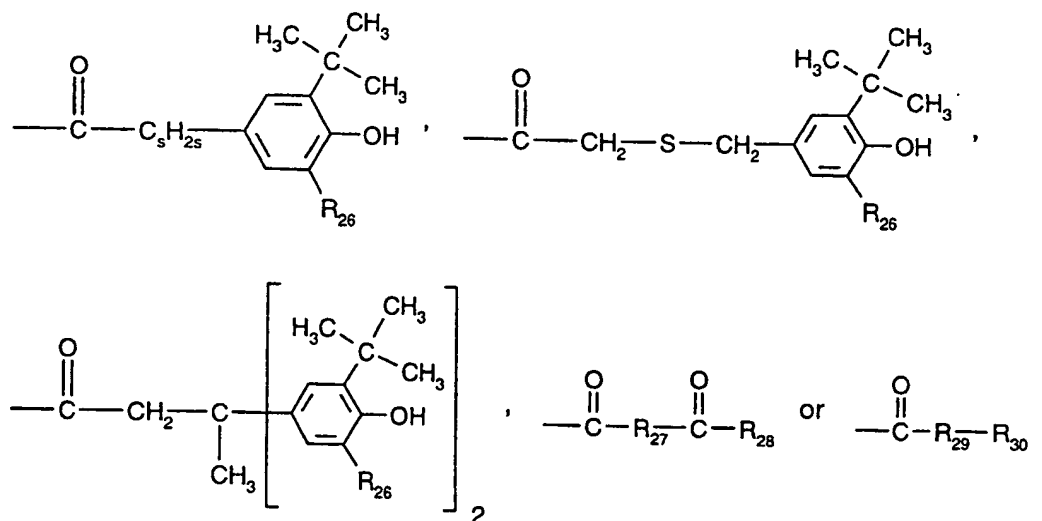
by from 1 to 3 $\text{C}_1\text{-C}_4$ alkyl groups, or furthermore the radicals R_{20} and R_{21} , together with the carbon atoms to which they are bonded, form a $\text{C}_5\text{-C}_{12}$ cycloalkylene ring unsubstituted or substituted by from 1 to 3 $\text{C}_1\text{-C}_4$ alkyl groups;

R_{22} is hydrogen or $\text{C}_1\text{-C}_4$ alkyl,

R_{23} is hydrogen, $\text{C}_1\text{-C}_{25}$ alkanoyl, $\text{C}_3\text{-C}_{25}$ alkenoyl; $\text{C}_3\text{-C}_{25}$ alkanoyl interrupted by oxygen, sul-

fur or by $\text{N}-\text{R}_{14}$; $\text{C}_2\text{-C}_{25}$ alkanoyl substituted by a di($\text{C}_1\text{-C}_6$ alkyl) phosphonate group;

$\text{C}_6\text{-C}_9$ cycloalkylcarbonyl, thenoyl, furoyl, benzoyl, or benzoyl substituted by $\text{C}_1\text{-C}_{12}$ alkyl;



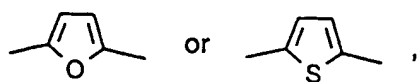
R_{24} and R_{25} are each independently of the other hydrogen or $\text{C}_1\text{-C}_{18}$ alkyl,

R_{26} is hydrogen or $\text{C}_1\text{-C}_8$ alkyl,

R_{27} is a direct bond, $\text{C}_1\text{-C}_{18}$ alkylene; $\text{C}_2\text{-C}_{18}$ alkylene interrupted by oxygen, sulfur or by

$\text{N}-\text{R}_{14}$; $\text{C}_2\text{-C}_{18}$ alkenylene, $\text{C}_2\text{-C}_{20}$ alkylidene, $\text{C}_7\text{-C}_{20}$ phenylalkylidene, $\text{C}_5\text{-C}_8$ cyclo-

alkylene, $\text{C}_7\text{-C}_8$ bicycloalkylene; phenylene unsubstituted or substituted by $\text{C}_1\text{-C}_4$ alkyl;



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R_{28} is hydroxy, $\left[-O^- \frac{1}{r} M^{r+} \right]$, C_1 - C_{18} alkoxy or $-N \begin{matrix} R_{24} \\ R_{25} \end{matrix}$,

R_{29} is oxygen, -NH- or $\begin{matrix} \diagup \\ N \end{matrix} - \overset{O}{\parallel} C - NH - R_{30} \begin{matrix} \diagdown \end{matrix}$,

R_{30} is C_1 - C_{18} alkyl or phenyl,

R_{31} is hydrogen or C_1 - C_{18} alkyl,

M is an r-valent metal cation,

X is a direct bond, oxygen, sulfur or $-NR_{31}-$,

n is 1 or 2,

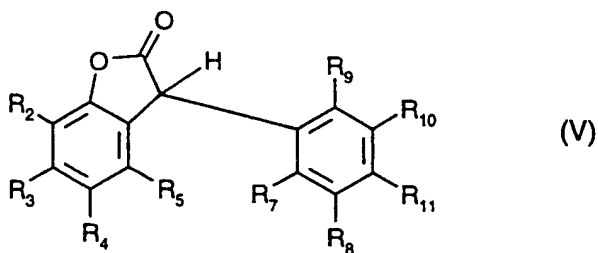
p is 0, 1 or 2,

q is 1, 2, 3, 4, 5 or 6,

r is 1, 2 or 3, and

s is 0, 1 or 2.

7. A composition according to claim 1, comprising as component (c) (i) a compound of formula V



wherein

R_2 is hydrogen or C_1 - C_6 alkyl,

R_3 is hydrogen,

R_4 is hydrogen or C_1 - C_6 alkyl,

R_5 is hydrogen,

R_7 , R_8 , R_9 , R_{10} and R_{11} are each independently of the others hydrogen, C_1 - C_4 alkyl, C_1 - C_4 -

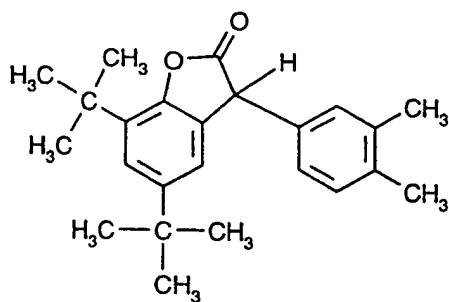
alkoxy or $\text{—O—}\begin{array}{c} R_{20} \\ | \\ \text{C} \\ | \\ \text{H} \end{array}\text{—}\begin{array}{c} R_{21} \\ | \\ \text{C} \\ | \\ R_{22} \end{array}\text{—O—}R_{23}$, with the proviso that at least two of the radicals R_7 ,

R_8 , R_9 , R_{10} and R_{11} are hydrogen,

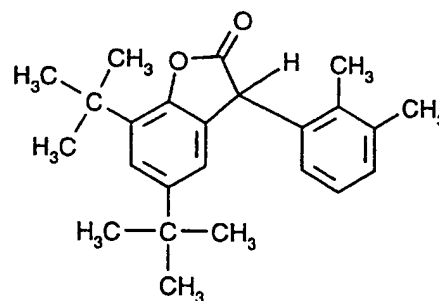
R_{20} , R_{21} and R_{22} are hydrogen, and

R_{23} is C_2 - C_4 alkanoyl.

8. A composition according to claim 1, wherein component (c) (i) is a compound of formula Va or Vb



(Va)



(Vb)

or a mixture of both compounds of formulae Va and Vb.

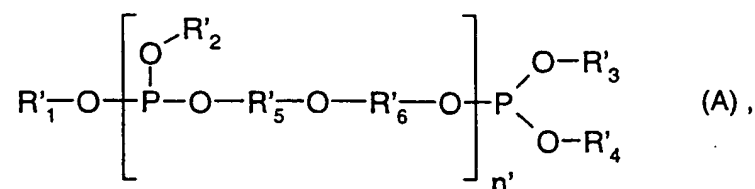
9. A composition according to claim 1, comprising as component (a) a lubricant, a fuel, a hydraulic fluid, a metal-working fluid or a synthetic polymer.

10. A composition according to claim 1, comprising as component (a) an engine oil, a turbine oil, a gear oil, a diesel fuel or a lubricating grease.

11. A composition according to claim 1, comprising as component (a) a synthetic polymer.

12. A composition according to claim 1, comprising as component (a) a polyolefin.

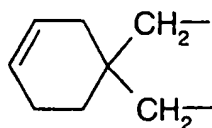
13. A composition according to claim 1, wherein component (b) is present in an amount of from 0.01 to 10 %, based on the weight of component (a).
14. A composition according to claim 1, wherein component (c) (i) is present in an amount of from 0.0005 to 5 %, based on the weight of component (a).
15. A composition according to claim 1, wherein component (c) (ii) is present in an amount of from 0.005 to 2 %, based on the weight of component (a).
16. A composition according to claim 1, comprising in addition, besides components (a), (b) and (c), further additives.
17. A composition according to claim 16, comprising as further additives phenolic antioxidants, light-stabilisers and/or processing stabilisers.
18. A stabiliser mixture comprising
 α) at least one compound of formula A



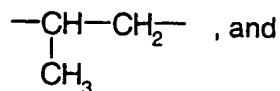
wherein

n' is a number from 1 to 6,

R'_1 , R'_2 , R'_3 and R'_4 are each independently of the others C_4 - C_{22} alkyl, C_7 - C_9 phenylalkyl; or C_5 - C_8 cycloalkyl unsubstituted or substituted by C_1 - C_4 alkyl; or R'_1 and R'_2 together and/or R'_3 and R'_4 together are C_2 - C_9 alkylene or a group of the formula



R'₅ and R'₆ are each independently of the other $-\text{CH}_2-\text{CH}_2-$ or



β) (i) at least one compound of the benzofuran-2-one type or (ii) α-tocopherol.

19. A stabiliser mixture according to claim 18, comprising, in addition, (γ) at least one compound from the group of phenolic antioxidants.

20. A stabiliser mixture according to claim 18, wherein the weight ratio of components (α) : (β) is from 100 : 0.01 to 0.01 : 100.

21. A stabiliser mixture according to claim 19, wherein the weight ratio of components (α) : (β) : (γ) is from 100 : 1 : 0.01 to 0.01 : 1 : 100.

22. A method of stabilising organic materials against oxidative, thermal or light-induced degradation wherein at least one each of components (b) and (c) according to claim 1 are incorporated therein or applied thereto.

23. The use of a mixture of components (b) and (c) according to claim 1 as stabilisers for organic materials against oxidative, thermal or light-induced degradation.

INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 99/01820

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 C09K15/32 C08K5/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C09K C08K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	GB 2 315 070 A (CIBA GEIGY AG) 21 January 1998 (1998-01-21) claims 1,3,7 ---	1-23
X	GB 2 252 325 A (CIBA GEIGY AG) 5 August 1992 (1992-08-05) claims 1,11 ---	1-23
P,X	GB 2 321 460 A (CIBA GEIGY AG) 29 July 1998 (1998-07-29) page 17; claim 1 ---	1-23
P,X	EP 0 839 623 A (CIBA GEIGY AG) 6 May 1998 (1998-05-06) claims 1,2 ---	1-23
	-/--	

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

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INTERNATIONAL SEARCH REPORT

Inter. Application No
PCT/EP 99/01820

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